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Phase-Conjugated Fluorescence

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Henk F. Arnoldus and Thomas F. George

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Fluorescent emission by an atom near a phase conjugator (PC) based on four-wave mixing is studied from first principles. The Maxwell-Heisenberg equations are solved for the radiation field, and with an asymptotic expansion an expression is derived for the field in the far zone. The total emitted power which can be measured by a detector in this region is evaluated, and it is found that this power acquires three distinct contributions. First, there are photons which are emitted by the atom directly towards the detector, and without any interaction with the medium. Second, there are photons that first travel towards the surface of the PC, and they have a certain probability of being reflected in the specular direction and towards the detector. The third kind of radiation consists of phase-conjugated photons, which are emitted independently of the previous ones. It is shown that the first two processes are a result of simple atomic spontaneous decay, but that the emission of a phase-conjugated fluorescent photon involves a three-photon process. The latter process has a probability proportional to the population of the atomic ground state. It is pointed out
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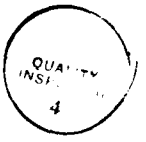

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PHASE-CONJUGATED FLUORESCENCE

Henk F. Arnoldus
Department of Physics
Mendel Hall
Villanova University
Villanova, Pennsylvania 19085

and

Thomas F. George
Departments of Chemistry and Physics & Astronomy
239 Fronczak Hall
State University of New York at Buffalo
Buffalo, New York 14260

Fluorescent emission by an atom near a phase conjugator (PC) based on four-wave mixing is studied from first principles. The Maxwell-Heisenberg equations are solved for the radiation field, and with an asymptotic expansion an expression is derived for the field in the far zone. The total emitted power which can be measured by a detector in this region is evaluated, and it is found that this power acquires three distinct contributions. First, there are photons which are emitted by the atom directly towards the detector, and without any interaction with the medium. Second, there are photons that first travel towards the surface of the PC, and they have a certain probability of being reflected in the specular direction and towards the detector. The third kind of radiation consists of phase-conjugated photons, which are emitted independently of the previous ones. It is shown that the first two processes are a result of simple atomic spontaneous decay, but that the emmission of a phase-conjugated fluorescent photon involves a three-photon process. The latter process has a probability proportional to the population of the atomic ground state. It is pointed out that an atom in its ground state polarizes the nonlinear medium of the PC, which subsequently can emit spontaneously two photons. An absorption-emission-absorption process by the atom then produces a fluorescent photon, together with a spontaneous excitation of the atom.

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I. INTRODUCTION

Wavefront inversion of optical radiation was observed for the first time by Zel'dovich *et al*[1] in stimulated Brillouin scattering. Mainly due to the fact that wavefront inversion, or phase conjugation, can be utilized to correct wavefront distortions in optical amplifiers,[2,3] this technique has since attracted a considerable amount of attention. In 1977 it was proposed by Hellwarth[4] and Yariv and Pepper[5] to construct a phase conjugator (PC) which operates through four-wave mixing (FWM) in a nonlinear medium, and almost simultaneously this was realized experimentally in a liquid CS_2 cell[6,7] and in a lithium formate crystal.[8] Alternative methods of phase conjugation include FWM in gases[9-11] and thin films,[12,13] or more exotic media like microparticles suspended in a liquid[14] or organic dye molecules in a solid matrix.[15] Also, the change in reflectivity of a thin metal layer under high irradiance has been considered as a candidate for the generation of phase-conjugated light.[16,17] More recently, phase conjugation in photorefractive crystals, like $BaTiO_3$, has gained popularity,[18] which can be attributed to the possibility of self-pumping of the crystal by the incident beam whose phase-conjugated image is sought.[19-20] On the theoretical side, much effort has been devoted to the description of (idealized) phase-conjugated radiation, and to the study of the applicability of a PC for wavefront correction.[21-25]

The possibility of wavefront distortion correction after phase conjugation relies on the equivalence between phase conjugation and time reversal, as explained in various reviews.[26-29] Taking the complex conjugate of the spatial part (the phase) of a field is mathematically equivalent to replacing t by -t. Many of the properties of phase-conjugated radiation can be understood most easily with time-reversal arguments. For instance, when a plane wave is incident on a phase-conjugating crystal (under a certain angle),

then the time-reversed replica of this beam must again be a plane wave, and it propagates in the direction opposite to the incident wave. This is in contrast to specular reflection at a linear medium (dielectric or metal). Also, when a point source of radiation, like a fluorescing atom, is close to the surface of a PC, then the incident field is a diverging spherical wave. According to the time-reversal argument, the reflected (phase-conjugated) wave must be a converging spherical wave which is focused exactly on the point source. This reflected field at the position of the source can affect the dynamical evolution of the radiatior considerably, and thereby its mechanism of emission, as was realized for the first time by Agarwal.[30]

Recently, Bochove[31] has redone the calculations of Ref. 30, with an approach where the radiation field is quantized explicitly. In both references the Einstein coefficient for spontaneous decay of an oscillating dipole was obtained, and it appeared indeed that there are some remarkable differences as compared to fluorescent emission in empty space or near a metal surface. Most notably, it was found that the decay rate is independent of the distance between the dipole and the PC, as could be expected from the equivalence of phase conjugation and time reversal. A reflected photon returns to the atoms, no matter how far away this atom is. Of course, there are some limitations on this result due to causality requirements and retardation effects, as recognized by the authors. Recently, Cook and Milonni[32] have shown that a sample of many two-level atoms in their ground states and near a PC is unstable. We shall show that this result also holds for a single atom, and explain the underlying physical mechanism. Hendricks and Nienhuis[33] have also studied the spontaneous decay of a two-level atom near a PC. Their results for the decay rate are consistent with our findings for the emission rate.

We consider an atomic dipole $\underline{\mu}(t)$ with arbitrary time dependence and evaluate the radiation field in the far zone, as it can be observed experimentally by a photodetector. The time dependence of the Heisenberg operator $\underline{\mu}(t)$ is brought about by its interaction with all components of the electric field. We were able to avoid an explicit quantization in terms of plane-wave modes of the radiation field, and the general results do not depend in form on the specific properties of the dipole moment $\underline{\mu}(t)$. We work out the case of a two-level atom, and it appears that some quite remarkable features in the behavior of an atom near a PC can be predicted without knowledge of the details of the temporal evolution of the atomic density operator.

II. VARIOUS FIELD COMPONENTS

A nonlinear medium occupies a part of the region $z < 0$, and its surface is the plane $z = 0$. Two counterpropagating laser beams with frequency $\bar{\omega} > 0$ pump the medium, and the third-order nonlinear susceptibility is responsible for a four-wave mixing process between the two pump fields, an incident field, and a generated phase-conjugated wave. We shall allow the medium to have a non-unity dielectric constant, which implies that we have to take into account the specular reflection of an incident wave. In this fashion we can keep track of the differences between linear and nonlinear effects, since they are both unified in a single formalism. A dipole $\underline{\mu}(t)$ is located at $\underline{r} = h\underline{e}_z$, $h > 0$, on the positive z-axis.

We are interested in the electric field operator $\underline{E}(\underline{r},t)$ in the region $z > 0$ only, since that is the place where we can put a detector and measure the radiation. In general, $\underline{E}(\underline{r},t)$ acquires three distinct contributions, and we can write

$$\underset{\sim}{E}(\underset{\sim}{r},t) = \underset{\sim}{E}_f(\underset{\sim}{r},t) + \underset{\sim}{E}_p(\underset{\sim}{r},t) + \underset{\sim}{E}_h(\underset{\sim}{r},t) \tag{2.1}$$

for the field in the Heisenberg picture. Here, $\underset{\sim}{E}_f$ is the free field which includes the vacuum field and possible external fields. The term $\underset{\sim}{E}_p$ is the particular solution and is equal to the radiation field of an atomic dipole in empty space. A part of this field is incident upon the PC, which generates a phase-conjugated signal and a reflected specular wave, both of which are included in $\underset{\sim}{E}_h$ (h = homogeneous solution).

The key to the evaluation of $\underset{\sim}{E}(\underset{\sim}{r},t)$ is the notion that the Heisenberg equations for the time evolution of the electromagnetic-field operators are identical in form to the classical Maxwell's equations.[34] Just as in classical electrodynamics, we cannot consider the field in the region $z > 0$ only, but we have to take into account the solution in $z < 0$ as well. At the boundary the two solutions must match in the usual way. The free field $\underset{\sim}{E}_f$ is assumed to obey Maxwell's equations separately. For parametric FWM, these equations are linear in the fields (in the undepleted-pump approximation), and therefore $\underset{\sim}{E}$-$\underset{\sim}{E}_f$ must also be a solution of Maxwell's equations. We call this the source field and write it as

$$\underset{\sim}{E}_s(\underset{\sim}{r},t) = \underset{\sim}{E}_p(\underset{\sim}{r},t) + \underset{\sim}{E}_h(\underset{\sim}{r},t) \quad , \tag{2.2}$$

for $z > 0$. This component of the radiation field is due to the presence of the dipole and its interaction with the medium. We shall focus our attention on $\underset{\sim}{E}_s$ only, since this is the radiative part which can be measured by a detector in the far zone. The homogeneous component has two distinct contributions,

$$\underline{E}_h(\underline{r},t) = \underline{E}_r(\underline{r},t) + \underline{E}_{pc}(\underline{r},t) \quad , \tag{2.3}$$

in terms of a specular (r) and a phase-conjugated (pc) part.

It is convenient to adopt a Fourier transform of $\underline{E}(\underline{r},t)$ according to

$$\hat{\underline{E}}(\underline{r},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\, \underline{E}(\underline{r},t) \quad . \tag{2.4}$$

The electric-field operator is Hermitian, which translates into the Fourier domain as

$$\underline{E}(\underline{r},t)^{\dagger} = \underline{E}(\underline{r},t) \longrightarrow \hat{\underline{E}}(\underline{r},\omega)^{\dagger} = \hat{\underline{E}}(\underline{r},-\omega) \quad . \tag{2.5}$$

A useful concept is the positive-frequency part of the field, which is defined as

$$\underline{E}(\underline{r},t)^{(+)} = \frac{1}{2\pi} \int_0^{\infty} d\omega\, e^{-i\omega t}\, \hat{\underline{E}}(\underline{r},\omega) \quad , \tag{2.6}$$

in terms of $\hat{\underline{E}}(\underline{r},\omega)$. The negative-frequency part follows from

$$\underline{E}(\underline{r},t)^{(-)} = [\underline{E}(\underline{r},t)^{(+)}]^{\dagger} \quad , \tag{2.7}$$

and the total field assumes the form

$$\underline{E}(\underline{r},t) = \underline{E}(\underline{r},t)^{(+)} + \underline{E}(\underline{r},t)^{(-)} \quad . \tag{2.8}$$

Similar notations will be used for other time-dependent quantities, and most notably for the dipole operator $\underset{\sim}{\mu}(t)$, which is also Hermitian. From Eq. (2.5) it follows that we know the field $\hat{\underset{\sim}{E}}(\underset{\sim}{r},\omega)$ as soon as its value for $\omega > 0$ only is given. Equivalently, Eqs. (2.7) and (2.8) show that the entire field is determined by just $\underset{\sim}{E}^{(+)}$ or $\underset{\sim}{E}^{(-)}$.

III. ANGULAR SPECTRUM OF PLANE WAVES

In this section we derive an explicit form of the particular solution $\underset{\sim}{E}_p$ for a dipole in empty space, which is most suitable for the evaluation of the homogeneous contribution (Sec. V). For $\omega > 0$, the Fourier transform of the electric field of a dipole at $\underset{\sim}{r} = \underset{\sim}{h}$ assumes the form

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = \frac{1}{4\pi\epsilon_0}\left(k^2\hat{\underset{\sim}{\mu}}(\omega) + (\hat{\underset{\sim}{\mu}}(\omega)\cdot\nabla)\nabla\right)\frac{e^{ik|\underset{\sim}{r}-\underset{\sim}{h}|}}{|\underset{\sim}{r}-\underset{\sim}{h}|} \quad , \tag{3.1}$$

with $k = \omega/c$, and where only $\hat{\underset{\sim}{\mu}}(\omega)$ is a quantum operator. After carrying out the differentiations, this expression reduces to the more familiar form.[35] We shall use Weyl's representation of the Green's function[36]

$$\frac{e^{ik|\underset{\sim}{r}-\underset{\sim}{h}|}}{|\underset{\sim}{r}-\underset{\sim}{h}|} = \frac{i}{2\pi}\int_{-\infty}^{\infty}d\alpha\int_{-\infty}^{\infty}d\beta\,\frac{1}{\gamma}\,e^{i\alpha x+i\beta y+i\gamma|z-h|} \quad , \quad k > 0 \ . \tag{3.2}$$

The parameter γ is defined by

$$\gamma = \begin{cases} \sqrt{k^2 - \alpha^2 - \beta^2} \\ \\ i\sqrt{\alpha^2 + \beta^2 - k^2} \end{cases} \ . \tag{3.3}$$

and we take the form for which the argument of the square root is positive.

Substituting expression (3.2) into Eq. (3.1) and carrying out the differentiations yields

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = -\frac{1}{\epsilon_o}\hat{\underset{\sim}{\mu}}(\omega)_\perp \delta(\underset{\sim}{r}-\underset{\sim}{h})$$

$$+\frac{i}{8\pi^2\epsilon_o}\int_{-\infty}^{\infty}d\alpha\int_{-\infty}^{\infty}d\beta\,\frac{1}{\gamma}\,e^{i\alpha x+i\beta y+i\gamma|z-h|}$$

$$\times\{k^2\hat{\underset{\sim}{\mu}}(\omega) - [\alpha\hat{\mu}_x(\omega)+\beta\hat{\mu}_y(\omega)+\gamma\,\mathrm{sgn}(z-h)\hat{\mu}_z(\omega)]$$

$$\times[\alpha\underset{\sim}{e}_x+\beta\underset{\sim}{e}_y+\gamma\,\mathrm{sgn}(z-h)\underset{\sim}{e}_z]\}\quad . \tag{3.4}$$

Here, $\hat{\underset{\sim}{\mu}}(\omega)_\perp$ is the perpendicular component of $\hat{\underset{\sim}{\mu}}(\omega)$ with respect to the plane $z = 0$. The δ-function at $\underset{\sim}{r} = \underset{\sim}{h}$ appears due to the second derivative of $|z-h|$ with respect to z. For two given values of the integration variables α and β, we define the vector

$$\underset{\sim}{K}_\parallel = \alpha\underset{\sim}{e}_x + \beta\underset{\sim}{e}_y \quad , \tag{3.5}$$

where the subscript $\parallel$ indicates that this vector is parallel to the $z = 0$ plane. In terms of γ, we then introduce the two complementary vectors

$$\underset{\sim}{K} = \underset{\sim}{K}_\parallel + \gamma\underset{\sim}{e}_z \quad , \tag{3.6}$$

$$\underset{\sim}{K}' = \underset{\sim}{K}_\parallel - \gamma\underset{\sim}{e}_z \quad , \tag{3.7}$$

which are each other's mirror image. Notice that $\underline{K}_{\parallel}$ is real, but that $\underline{K}$ and $\underline{K}'$ can each have an imaginary z-component. For the square of the magnitude of the various wave vectors, we find

$$K^2 = (K')^2 = k^2 = \alpha^2 + \beta^2 + \gamma^2 = (\omega/c)^2 \quad . \tag{3.8}$$

Furthermore, we write symbolically

$$\int d^2\underline{K}_{\parallel}(\ldots) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta \ (\ldots) \quad . \tag{3.9}$$

With the notations from above, $\hat{\underline{E}}_p(\underline{r},\omega)$ can be cast in the form

$$\hat{\underline{E}}_p(\underline{r},\omega) = \frac{i}{8\pi^2\epsilon_o} \int d^2\underline{K}_{\parallel} \frac{1}{\gamma} e^{i\underline{K}\cdot(\underline{r}-\underline{h})}$$

$$\times (k^2\hat{\underline{\mu}}(\omega) - (\hat{\underline{\mu}}(\omega)\cdot\underline{K})\underline{K}), \quad \text{for } z > h \quad , \tag{3.10}$$

and the expression for $0 < z < h$ follows from Eq. (3.10) after the substitution $\underline{K} \rightarrow \underline{K}'$ in the integrand.

For every value of $\underline{K}_{\parallel}$, the integrand of Eq. (3.10) represents a plane wave with wave vector $\underline{K}$, and because of the identity

$$\underline{K}\cdot(k^2\hat{\underline{\mu}}(\omega) - (\hat{\underline{\mu}}(\omega)\cdot\underline{K})\underline{K}) = 0 \quad , \tag{3.11}$$

every wave is transverse. The z-component of the wave vector $\underline{K}$ equals γ from Eq. (3.3), which is either positive or positive-imaginary, corresponding, respectively, to a wave which travels in the positive z-direction or to a wave

which decays exponentially in amplitude in the same direction (evanescent wave). For the field in the region $0 < z < h$, the wave vector is $\underset{\sim}{K}'$ for a given $\underset{\sim}{K}_{\parallel}$, and the wave is again either travelling or evanescent. The various occuring waves are illustrated pictorially in Fig. 1.

IV. POLARIZED WAVES

For reflection of a plane wave at a dielectric, the ratio of the amplitudes of the reflected field and the incident field, including the phase, is given by the appropriate Fresnel coefficient. This coefficient depends on the angle of incidence and the polarization of the incident wave. It can be shown[37] that the same holds for reflection of a phase-conjugated wave by a nonlinear medium. Therefore, it is advantageous to decompose the various plane waves in $\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega)$ into surface(s)- and plane(p)-polarized waves. For a given $\underset{\sim}{K}$ or $\underset{\sim}{K}'$ we define

$$\underset{\sim}{e}_{\underset{\sim}{K}s} = \underset{\sim}{e}_{\underset{\sim}{K}'s} = \frac{1}{K_{\parallel}} \underset{\sim}{K}_{\parallel} \times \underset{\sim}{e}_z \quad , \tag{4.1}$$

$$\underset{\sim}{e}_{\underset{\sim}{K}p} = -\frac{1}{kK_{\parallel}} (K_{\parallel}^2 \underset{\sim}{e}_z - \gamma \underset{\sim}{K}_{\parallel}) \quad , \tag{4.2}$$

$$\underset{\sim}{e}_{\underset{\sim}{K}'p} = -\frac{1}{kK_{\parallel}} (K_{\parallel}^2 \underset{\sim}{e}_z + \gamma \underset{\sim}{K}_{\parallel}) \quad . \tag{4.3}$$

These unit vectors are normalized according to $\underset{\sim}{e}_i \cdot \underset{\sim}{e}_i = 1$ for any subscript i, and they are perpendicular to their corresponding wave vector. Notice that the p-polarization vectors can have an imaginary z-component. The directions of the various unit vectors are depicted in Fig. 1. The field $\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega)$ in $z > h$ can then be decomposed as

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = \frac{i\omega^2}{8\pi^2\epsilon_o c^2}\sum_\sigma \int d^2\underset{\sim}{K}_\parallel \frac{1}{\gamma} e^{i\underset{\sim}{K}\cdot(\underset{\sim}{r}-\underset{\sim}{h})}(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\underset{\sim}{K}\sigma})\underset{\sim}{e}_{\underset{\sim}{K}\sigma} \quad , \tag{4.4}$$

with $\sigma = s,p$, and with $\underset{\sim}{K} \to \underset{\sim}{K}'$ we obtain the field for $0 < z < h$.

V. REFLECTED FIELDS

In order to simplify the notation, we introduce the two quantum operators

$$E_{\underset{\sim}{K}\sigma} = \frac{i\omega^2 e^{-ih\gamma}}{8\pi^2\epsilon_o c^2\gamma}(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\underset{\sim}{K}\sigma}) \quad . \tag{5.1}$$

$$E_{\underset{\sim}{K}'\sigma} = \frac{i\omega^2 e^{ih\gamma}}{8\pi^2\epsilon_o c^2\gamma}(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\underset{\sim}{K}'\sigma}) \quad . \tag{5.2}$$

Then the particular solution can be written as

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = \sum_\sigma \int d^2\underset{\sim}{K}_\parallel E_{\underset{\sim}{K}\sigma}\underset{\sim}{e}_{\underset{\sim}{K}\sigma} e^{i\underset{\sim}{K}\cdot\underset{\sim}{r}} \quad , \qquad \text{for } z > h \quad , \tag{5.3}$$

and with $\underset{\sim}{K} \to \underset{\sim}{K}'$ we find the field in the region $0 < z < h$. Expression (5.3) with $\underset{\sim}{K} \to \underset{\sim}{K}'$ is a superposition of polarized plane waves, where $E_{\underset{\sim}{K}'\sigma}$ serves as the amplitude, including the proper phase. Since Maxwell's equations are linear, we can calculate the reflected field on a per-wave basis. In the region $0 < z < h$, each plane wave has the form $E_{\underset{\sim}{K}'\sigma}\underset{\sim}{e}_{\underset{\sim}{K}'\sigma}\exp(i\underset{\sim}{K}'\cdot\underset{\sim}{r})$, and these waves are the incident waves on the medium (Fig. 1), which have to be matched across the boundary $z = 0$ to a solution in $z < 0$.

When a plane wave with wave vector $\underset{\sim}{K}'$, frequency $\omega > 0$ and polarization σ is incident upon the medium, then the reflected radiation consists again of σ-polarized plane waves, but with different wave vectors, in general. Due to the four-wave mixing, the frequency of the reflected wave can also be different. When the two pumps have frequency $\bar{\omega} > 0$, and the incident wave has frequency $\omega > 0$, then it can be shown[37] that the FWM process couples the field with frequency ω to the field with frequency

$$\omega' = \omega - 2\bar{\omega} \quad , \tag{5.4}$$

which is negative. Therefore, reflected radiation can be generated in the medium, both at frequency ω and at frequency ω', but not at other frequencies.

At the interface $z = 0$, the spatial part of the incident wave reduces to $\exp(i\underset{\sim}{K}_{\parallel} \cdot \underset{\sim}{r})$. It is only possible to match this wave to other waves for all $\underset{\sim}{r}$ simultaneously when the other waves have the same spatial dependence. Therefore, all waves inside and outside the medium must have a wave vector with the same parallel component $\underset{\sim}{K}_{\parallel}$. For the wave vector of the reflected wave at frequency ω, we can write $\underset{\sim}{K}'_r = \underset{\sim}{K}_{\parallel} + K'_{r,z}\, \underset{\sim}{e}_z$, for which it must hold that $(K'_{r,z})^2 = \omega^2/c^2 - K_{\parallel}^2 = \gamma^2$. The solution $K'_{r,z} = -\gamma$ corresponds to the incident wave, and the only other possibility is $K'_{r,z} = +\gamma$. But this is the z-component of the complementary wave $\underset{\sim}{K}$, so that we find $\underset{\sim}{K}_r = \underset{\sim}{K}$. Figure 1 shows that this wave is just the ordinary specular wave, although its generation is affected by the nonlinear interaction. For the wave at frequency ω' we write

$$\underset{\sim}{K}'_{pc} = \underset{\sim}{K}_{\parallel} + K'_{pc,z}\, \underset{\sim}{e}_z \quad , \tag{5.5}$$

where

$$K'_{pc,z} = \begin{cases} -\sqrt{(\omega'/c)^2 - K_\parallel^2} \\ \\ i\sqrt{K_\parallel^2 - (\omega'/c)^2} \end{cases} \quad . \tag{5.6}$$

In general, we shall have $|\omega'| \approx |\omega|$, which gives $K'_{pc,z} \approx -\gamma$ for the case of a travelling wave, and thereby $\underset{\sim}{K}'_{pc} \approx \underset{\sim}{K}'$. This shows that the incident wave and the pc-wave are approximately counterpropagating, as is required for a time-reversed wave.

Next we have to choose a phase convention for the unit polarization vectors of the r-wave and the pc-wave. Since $\underset{\sim}{K}'_r = \underset{\sim}{K}$, we can simply take

$$\underset{\sim}{e}_{\underset{\sim}{K}'_r \sigma} = \underset{\sim}{e}_{\underset{\sim}{K}\sigma} \quad , \qquad \sigma = s,p \quad , \tag{5.7}$$

for the r-wave, where the right-hand side is given by Eqs. (4.1) and (4.2). A convenient choice for the pc-wave is

$$\underset{\sim}{e}_{\underset{\sim}{K}'_{pc} s} = \frac{1}{K_\parallel} \underset{\sim}{K}_\parallel \times \underset{\sim}{e}_z \quad , \tag{5.8}$$

$$\underset{\sim}{e}_{\underset{\sim}{K}'_{pc} p} = - \frac{c}{\omega'} \underset{\sim}{K}'_{pc} \times \underset{\sim}{e}_{\underset{\sim}{K}'_{pc} s} \quad , \tag{5.9}$$

which are both normalized according to $\underset{\sim}{e}_i \cdot \underset{\sim}{e}_i = 1$, and are both perpendicular to $\underset{\sim}{K}'_{pc}$.

For a given incident field of the form (5.3) with $\underset{\sim}{K} \rightarrow \underset{\sim}{K}'$, the reflected field (homogeneous solution) has two components. The r-wave at frequency ω attains the form

$$\hat{\underset{\sim}{E}}_r(\underset{\sim}{r},\omega) = \sum_\sigma \int d^2\underset{\sim}{K}_\parallel \; E_{\underset{\sim}{K}'\sigma} R_{\underset{\sim}{K}'\sigma} \underset{\sim}{e}_{\underset{\sim}{K}\sigma} e^{i\underset{\sim}{K}\cdot\underset{\sim}{r}} \quad , \tag{5.10}$$

and for the pc-wave we can write

$$\hat{\underset{\sim}{E}}_{pc}(\underset{\sim}{r},\omega') = \sum_\sigma \int d^2\underset{\sim}{K}_\parallel \; E_{\underset{\sim}{K}'\sigma} P_{\underset{\sim}{K}'\sigma} \underset{\sim}{e}_{\underset{\sim}{K}'_{pc}\sigma} e^{i\underset{\sim}{K}'_{pc}\cdot\underset{\sim}{r}} \quad . \tag{5.11}$$

Here, $R_{\underset{\sim}{K}'\sigma}$ and $P_{\underset{\sim}{K}'\sigma}$ are the Fresnel coefficients for reflection of the r-wave and the pc-wave, respectively. They depend in a complicated way on the properties of the medium and the details of the FWM, like the dielectric constant, the third-order susceptibility, the pump polarization, etc.

VI. TIME DOMAIN

Of practical interest is the electric field as a function of time. It is sufficient to evaluate the positive- or negative-frequency part only since these components determine the field completely. For the particular solution $\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega)$ we have Eq. (5.3) for $z > h$, which holds for $\omega > 0$ only. Therefore, the positive-frequency part of $\underset{\sim}{E}_p(\underset{\sim}{r},t)$ follows from Eq. (2.6), and is explicitly

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = \frac{1}{2\pi} \int_0^\infty d\omega \; e^{-i\omega t} \sum_\sigma \int d^2\underset{\sim}{K}_\parallel \; E_{\underset{\sim}{K}\sigma} \underset{\sim}{e}_{\underset{\sim}{K}\sigma} e^{i\underset{\sim}{K}\cdot\underset{\sim}{r}} \quad , \tag{6.1}$$

for $z > h$, and with $\underset{\sim}{K} \to \underset{\sim}{K}'$ we obtain the field in $0 < z < h$. In a similar way, the superposition of all specular waves has the representation

$$\underset{\sim}{E}_r(\underset{\sim}{r},t)^{(+)} = \frac{1}{2\pi}\int_0^\infty d\omega\, e^{-i\omega t}\sum_\sigma \int d^2\underset{\sim}{K}_\parallel\, E_{K'\sigma}{}^R{}_{\underset{\sim}{K}'\sigma}\,\underset{\sim}{e}_{\underset{\sim}{K}\sigma}\, e^{i\underset{\sim}{K}\cdot\underset{\sim}{r}} \quad . \tag{6.2}$$

For the phase-conjugated wave we have expression (5.11) in which the frequency ω' is negative. Consequently, a Fourier synthesis yields the negative-frequency part of the field. We obtain

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = \frac{1}{2\pi}\int_{-\infty}^0 d\omega'\, e^{-i\omega' t}\sum_\sigma \int d^2\underset{\sim}{K}_\parallel\, E_{\underset{\sim}{K}'\sigma}{}^P{}_{\underset{\sim}{K}'\sigma}\,\underset{\sim}{e}_{\underset{\sim}{K}'_{pc}\sigma}\, e^{i\underset{\sim}{K}'_{pc}\cdot\underset{\sim}{r}} \quad . \tag{6.3}$$

VII. ASYMPTOTIC EXPANSION

Although expressions (6.1)-(6.3) give the total source field in $z > 0$ as a function of time, the appearance of the integrals over the frequency and the parallel components of the wave vectors is cumbersome. Fortunately, for the study of fluorescent emission we do not need the exact solution for the radiation field at every point $\underset{\sim}{r}$ in the half-space $z > 0$. Only the value of $\underset{\sim}{E}_s(\underset{\sim}{r},t)$ for $r = |\underset{\sim}{r}| \to \infty$ is of relevance. In this section we evaluate the asymptotic behavior for $r \to \infty$, and in the next two sections we work out the formal results in order to obtain more managable and transparent expressions.

Suppose a detector is located at position $\underset{\sim}{r}$. This point will be represented by its spherical coordinates (r,θ,ϕ) with respect to the z-axis, and we are interested in the value of the radiation field for $r \to \infty$ with (θ,ϕ) fixed, and for $z > 0$ only. The integrals in Eqs. (6.1)-(6.3) have the form $\int d^2\underset{\sim}{K}_\parallel\, \exp(i\underset{\sim}{\kappa}\cdot\underset{\sim}{r})g(\underset{\sim}{K}_\parallel)$, with $\underset{\sim}{\kappa} = \underset{\sim}{K}$ or $\underset{\sim}{\kappa} = \underset{\sim}{K}'_{pc}$. With the method of stationary

phase,[38] we can derive an asymptotic expansion for integrals of this kind. For the p-wave and r-wave we obtain

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = -\frac{i\cos\theta}{rc}\int_0^\infty d\omega\, e^{-i\omega(t-r/c)}\,\omega\sum_\sigma [E_{\underset{\sim}{K}\sigma}\underset{\sim}{e}_{\underset{\sim}{K}\sigma}]_{\underset{\sim}{K}_\parallel=k(\underset{\sim}{e}_r)_\parallel}\quad , \tag{7.1}$$

$$\underset{\sim}{E}_r(\underset{\sim}{r},t)^{(+)} = -\frac{i\cos\theta}{rc}\int_0^\infty d\omega\, e^{-i\omega(t-r/c)}\,\omega\sum_\sigma [R_{\underset{\sim}{K}'\sigma}E_{\underset{\sim}{K}'\sigma}\underset{\sim}{e}_{\underset{\sim}{K}\sigma}]_{\underset{\sim}{K}_\parallel=k(\underset{\sim}{e}_r)_\parallel}\quad . \tag{7.2}$$

With

$$\rho = -\frac{\omega'}{\omega} = 1 + 2\frac{\bar{\omega}-\omega}{\omega}\quad , \tag{7.3}$$

the asymptotic expansion of the pc-wave becomes

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = -\frac{i\cos\theta}{rc}\int_{-\infty}^0 d\omega'\, e^{-i\omega'(t-r/c)}\,\omega'\sum_\sigma [P_{\underset{\sim}{K}'\sigma}E_{\underset{\sim}{K}'\sigma}\underset{\sim}{e}_{\underset{\sim}{K}'_{pc}\sigma}]_{\underset{\sim}{K}_\parallel=-\rho k(\underset{\sim}{e}_r)_\parallel}\quad . \tag{7.4}$$

The parameter ρ accounts for the mismatch between ω (or $-\omega'$) and the pump frequency $\bar{\omega}$.

VIII. AMPLITUDES

In the formal expansions (7.1), (7.2) and (7.4) we still have to evaluate the factors in square brackets for the indicated values of $\underset{\sim}{K}_\parallel$. In this section we consider the (quantum) amplitude factors $E_{\underset{\sim}{K}\sigma}$ and $E_{\underset{\sim}{K}'\sigma}$, as

by Eqs. (5.1) and (5.2), respectively. These factors contain the parameter γ, which depends on $K_\parallel^2 = \alpha^2 + \beta^2$ according to Eq. (3.3). We find

$$K_\parallel^2 = \begin{cases} k^2 \sin^2\vartheta \ , & \text{for the p- and r-wave} \\ \\ \rho^2 k^2 \sin^2\vartheta \ , & \text{for the pc-wave} \end{cases} , \tag{8.1}$$

which gives

$$\gamma = \begin{cases} k\cos\vartheta \ , & \text{for the p- and r-wave} \\ \\ k/\sqrt{1 - \rho^2 \sin^2\vartheta} \ , & \text{for the pc-wave} \end{cases} . \tag{8.2}$$

Substitution of $E_{\underset{\sim}{K}\sigma}$ and $E_{\underset{\sim}{K}',\sigma}$ into the expansions (7.1), (7.2) and (7.4) then gives for the three fields

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = \frac{1}{8\pi^2 \epsilon_o r c^2} \int_0^\infty d\omega \ e^{-i\omega(t-r/c+\tau)} \ \omega^2$$

$$\times \sum_\sigma \left[\underset{\sim}{e}_{\underset{\sim}{K}\sigma} (\hat{\underset{\sim}{\mu}}(\omega) \cdot \underset{\sim}{e}_{\underset{\sim}{K}\sigma}) \right]_{\underset{\sim}{K}_\parallel = k(\underset{\sim}{e}_r)_\parallel} , \tag{8.3}$$

$$\underset{\sim}{E}_r(\underset{\sim}{r},t)^{(+)} = \frac{1}{8\pi^2 \epsilon_o r c^2} \int_0^\infty d\omega \ e^{-i\omega(t-r/c-\tau)} \ \omega^2$$

$$\times \sum_\sigma \left[R_{\underset{\sim}{K}',\sigma} \underset{\sim}{e}_{\underset{\sim}{K}\sigma} (\hat{\underset{\sim}{\mu}}(\omega) \cdot \underset{\sim}{e}_{\underset{\sim}{K}',\sigma}) \right]_{\underset{\sim}{K}_\parallel = k(\underset{\sim}{e}_r)_\parallel} , \tag{8.4}$$

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = \frac{1}{8\pi^2\epsilon_o rc^2}\int_{-\infty}^{0} d\omega' e^{-i\omega'(t-r/c+r\xi/\rho)}\,\frac{\omega\omega'}{\xi}$$

$$\times \sum_{\sigma}\left[P_{\underset{\sim}{K}',\sigma}\underset{\sim}{e}_{\underset{\sim}{K}',pc\sigma}(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\underset{\sim}{K}',\sigma})\right]_{\underset{\sim}{K}_{\parallel}=-\rho k(\underset{\sim}{e}_r)_{\parallel}} , \tag{8.5}$$

where we introduced the parameters

$$\xi = \frac{\sqrt{1-\rho^2\sin^2\vartheta}}{\cos\vartheta} , \tag{8.6}$$

$$r = \frac{h}{c}\cos\vartheta . \tag{8.7}$$

At this stage it is convenient, although not necessary, to make a slight approximation in the expression for the phase-conjugated wave. For a given frequency ω', the integrand in Eq. (8.5) is proportional to the Fresnel reflection coefficient $P_{\underset{\sim}{K}',\sigma}$, which depends on ω'. It is well known[37] that $P_{\underset{\sim}{K}',\sigma}$ assumes only a finite value when the frequency ω of the incident wave is sufficiently close to the pump frequency $\bar{\omega}$. Since the integrand in Eq. (8.5) is proportional to $P_{\underset{\sim}{K}',\sigma}$, we can effectively set $\rho = 1$ in any factor that multiplies $P_{\underset{\sim}{K}',\sigma}$. Similarly, we can set $\xi = 1$ and $\omega' = -\omega$ in the integrand, and also $\underset{\sim}{e}_{\underset{\sim}{K}',pc\sigma} = \underset{\sim}{e}_{\underset{\sim}{K}',\sigma}$ as can be verified by inspection of the definition of these unit vectors. Then Eq. (8.5) reduces to

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = -\frac{1}{8\pi^2\epsilon_o rc^2}\int_{-\infty}^{0} d\omega'\; e^{-i\omega'(t-r/c+r)}\;\omega^2$$

$$\times \sum_{\sigma}\left[P_{\underset{\sim}{K}',\sigma}\underset{\sim}{e}_{\underset{\sim}{K}',\sigma}(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\underset{\sim}{K}',\sigma})\right]_{\underset{\sim}{K}_{\parallel}=-k(\underset{\sim}{e}_r)_{\parallel}} , \tag{8.8}$$

which is a great simplification. Notice that we have set $\rho = 1$ only in the overall factors, but the full resonant frequency dependence is retained, as it is incorporated in $P_{\underset{\sim}{K}'\sigma}$.

IX. POLARIZATION VECTORS, FRESNEL COEFFICIENTS AND THE MIRROR DIPOLE

Next we have to evaluate the polarization vectors $\underset{\sim}{e}_{\underset{\sim}{K}\sigma}$ and $\underset{\sim}{e}_{\underset{\sim}{K}'\sigma}$, as they are defined by Eqs. (4.1)-(4.3), and for the indicated values of $\underset{\sim}{K}_{\parallel}$ in Eqs. (8.3), (8.4) and (8.8). The parameter γ in Eqs. (4.1)-(4.3) equals $\gamma = k\cos\theta$ in all cases, as follows from Eq. (8.2) with $\rho \to 1$. We wish to express the polarization vectors in the standard spherical unit vectors $\underset{\sim}{e}_{\vartheta}$ and $\underset{\sim}{e}_{\phi}$ for a given observation angle (ϑ,ϕ).

For the particular solution (p-wave) from Eq. (8.3) we need $\underset{\sim}{e}_{\underset{\sim}{K}\sigma}$ for $\underset{\sim}{K}_{\parallel} = k(\underset{\sim}{e}_r)_{\parallel}$, both for $\sigma = s$ and $\sigma = p$. We find

$$[\underset{\sim}{e}_{\underset{\sim}{K}s}]_{\underset{\sim}{K}_{\parallel}=k(\underset{\sim}{e}_r)_{\parallel}} = -\underset{\sim}{e}_{\phi} \quad , \tag{9.1}$$

$$[\underset{\sim}{e}_{\underset{\sim}{K}p}]_{\underset{\sim}{K}_{\parallel}=k(\underset{\sim}{e}_r)_{\parallel}} = \underset{\sim}{e}_{\vartheta} \quad , \tag{9.2}$$

as could be expected from a comparison between the phase conventions for $\underset{\sim}{e}_{\underset{\sim}{K}s}$ and $\underset{\sim}{e}_{\underset{\sim}{K}p}$ (Fig. 1), and the definitions of $\underset{\sim}{e}_{\phi}$ and $\underset{\sim}{e}_{\vartheta}$, respectively. The field then attains the form

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = \frac{1}{8\pi^2\epsilon_o rc^2}\int_0^\infty d\omega\, e^{-i\omega(t-r/c+\tau)}\,\omega^2$$

$$\times\,[(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\vartheta})\underset{\sim}{e}_{\theta} + (\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\phi})\underset{\sim}{e}_{\phi}] \quad , \tag{9.3}$$

fluorescence, which is of course unphysical. It can be shown,[41] however, that when we allow for a finite but very small detuning $\omega_o - \bar{\omega} \neq 0$, the Fresnel coefficients remain finite, with a magnitude on the order of unity. This renders finite values for the parameters $c_{\parallel}$ and $c_{\perp}$, and thereby for the emission rate. For an extremely close resonance between ω_o and $\bar{\omega}$ we can no longer replace $P_\sigma(\omega,\cos\theta)$ in Eq. (9.15) by $P_\sigma(\omega_o,\cos\theta)$, and we have to take into account the frequency dependence of $P_\sigma(\omega,\cos\theta)$ over the width of the atomic emission line.

This situation is analogous to the problem of resonance fluorescence by a two-level atom in a monochromatic laser field. There, the spectral distribution of the fluorescence consists of a δ-function at the laser frequency, superposed on a smooth background, and in the limit where ω_o equals the laser frequency the emission rate is infinite. In any case, the example of this section shows that the situation of perfect resonance has to be considered with caution.

XV. SCHRÖDINGER PICTURE

The time dependence of the emission rate dW/dt in Eq. (13.6) is governed by the time evolution of the Heisenberg operator $\underline{\mu}(t)$. A more transparent representation of dW/dt can be obtained by a transformation to the Schrödinger picture. When we take the Schrödinger and Heisenberg picture to coincide at $t = 0$, then we have

$$\underline{\mu}(0) = \underline{\mu} = \underline{\mu}^{(+)} + \underline{\mu}^{(-)} \quad , \tag{15.1}$$

with

and notice that the factor in brackets is just the transverse component of $\hat{\underline{\mu}}(\omega)$ with respect to the propagation direction $\underline{e}_r$.

For the r-wave from Eq. (8.4) we need again $\underline{e}_{\underline{K}\sigma}$ at $\underline{K}_\parallel = k(\underline{e}_r)_\parallel$ for the unit polarization vectors which correspond to a propagating wave with wave vector $\underline{K}$ in the observation direction (θ,ϕ). In addition, we need $\underline{e}_{\underline{K}'\sigma}$ at the same value of $\underline{K}_\parallel$, and these unit vectors account for the polarization of an incident wave on the surface $z = 0$, which is subsequently emitted in the specular direction (Fig. 1). We find

$$\underline{e}_{\underline{K}'s} \cdot \underline{K}_\parallel = k(\underline{e}_r)_\parallel = -\underline{e}_\phi \quad , \tag{9.4}$$

$$\underline{e}_{\underline{K}'p} \cdot \underline{K}_\parallel = k(\underline{e}_r)_\parallel = -\underline{e}_\theta - 2\sin\theta \underline{e}_z \quad , \tag{9.5}$$

where the term $-2\sin\theta\underline{e}_z$ represents the change in direction between $\underline{K}'$ and $\underline{K}$ at the moment of reflection at the interface $z = 0$. For s-polarized waves there is no such correction, since both the incident wave and the specular have the same polarization vector (Eq. (4.1)). The specular field becomes

$$\underline{E}_r(\underline{r},t)^{(+)} = \frac{1}{8\pi^2 \epsilon_o r c^2} \int_0^\infty d\omega \, e^{-i\omega(t-r/c-\tau)} \, \omega^2$$

$$\times \left(R_{\underline{K}'p} (\hat{\underline{\mu}}(\omega) \cdot (-\underline{e}_\theta - 2\sin\theta \underline{e}_z)) \underline{e}_\theta + R_{\underline{K}'s} (\hat{\underline{\mu}}(\omega) \cdot \underline{e}_\phi) \underline{e}_\phi \right) \quad , \tag{9.6}$$

where the Fresnel coefficients $R_{\underline{K}'\sigma}$ pertain to the reflection coefficients of incident waves with wave vectors $\underline{K}'$, which are specularly reflected in the observation direction (θ,ϕ). The coefficients $R_{\underline{K}'\sigma}$ depend on the frequency ω

and angle of incidence θ_i. Since θ_i equals the observation angle θ, we might as well label $R_{\underline{K}',\sigma}$ with θ, rather than θ_i. Therefore, we can write

$$R_{\underline{K}',\sigma} = R_\sigma(\omega,\cos\theta) \quad , \quad \sigma = s,p \quad . \tag{9.7}$$

A convenient concept for specular reflection is the mirror dipole. If we decompose $\underline{\mu}$ (or $\hat{\underline{\mu}}$) as

$$\underline{\mu} = \underline{\mu}_\perp + \underline{\mu}_\parallel \quad , \tag{9.8}$$

where the subscripts $\perp$ and $\parallel$ refer to the surface $z = 0$, then the mirror dipole is defined as

$$\underline{\mu}' = \underline{\mu}_\perp - \underline{\mu}_\parallel \quad . \tag{9.9}$$

Combining everything then yields for the specular field

$$\underline{E}_r(\underline{r},t)^{(+)} = \frac{1}{8\pi^2\epsilon_o rc^2}\int_0^\infty d\omega\, e^{-i\omega(t-r/c-\tau)}\,\omega^2$$

$$\times \{R_p(\omega,\cos\theta)(\hat{\underline{\mu}}'(\omega)\cdot\underline{e}_\theta)\underline{e}_\theta$$

$$- R_s(\omega,\cos\theta)(\hat{\underline{\mu}}'(\omega)\cdot\underline{e}_\phi)\underline{e}_\phi\} \quad . \tag{9.10}$$

The minus sign in front of R_s is a consequence of the phase convention of the polarization vectors for s-waves.

For the phase-conjugated field we have to calculate $\underline{e}_{\underline{K}',\sigma}$ at $\underline{K}_\parallel$ =

$-k(\underset{\sim}{e}_r)_{\parallel}$. Both unit vectors in Eq. (8.8) appear with wave vector $\underset{\sim}{K}'$, which reflects the fact that both the incident plane wave and the pc-wave have the same wave vector $\underset{\sim}{K}'$. The additional minus sign in $\underset{\sim}{K}_{\parallel}$, as compared to $\underset{\sim}{K}_{\parallel}$ for the p-wave and the r-wave, signifies that a phase-conjugated plane wave travels in the direction opposite to its wave vector. The polarization vectors are

$$[\underset{\sim}{e}_{\underset{\sim}{K}'s}]_{\underset{\sim}{K}_{\parallel}=-k(\underset{\sim}{e}_r)_{\parallel}} = \underset{\sim}{e}_{\phi} \quad , \tag{9.11}$$

$$[\underset{\sim}{e}_{\underset{\sim}{K}'p}]_{\underset{\sim}{K}_{\parallel}=-k(\underset{\sim}{e}_r)_{\parallel}} = \underset{\sim}{e}_{\vartheta} \quad , \tag{9.12}$$

and the field is found to be

$$\begin{aligned} \underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = & -\frac{1}{8\pi^2\epsilon_o rc^2}\int_{-\infty}^{0} d\omega'\, e^{-i\omega'(t-r/c+\tau)}\,\omega^2 \\ & \times \{P_p(\omega,\cos\theta)(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\vartheta})\underset{\sim}{e}_{\theta} \\ & + P_s(\omega,\cos\theta)(\hat{\underset{\sim}{\mu}}(\omega)\cdot\underset{\sim}{e}_{\phi})\underset{\sim}{e}_{\phi}\} \quad , \end{aligned} \tag{9.13}$$

where we introduced the notation

$$P_{\sigma}(\omega,\cos\theta) = P_{\underset{\sim}{K}'\sigma} \quad , \quad \sigma = s,p \quad . \tag{9.14}$$

A complication with expression (9.13) is that the integration runs over ω', whereas the factor in brackets depends on ω. With the relation $\omega' = \omega - 2\bar{\omega}$ we can write the field in the alternative form

$$\underline{E}_{pc}(\underline{r},t)^{(-)} = -\frac{1}{8\pi^2\epsilon_o rc^2} e^{2i\bar{\omega}(t-r/c+\tau)} \int_0^\infty d\omega\, e^{-i\omega(t-r/c+\tau)}\, \omega^2$$

$$\times \{P_p(\omega,\cos\theta)(\hat{\underline{\mu}}(\omega)\cdot\underline{e}_\theta)\underline{e}_\theta$$

$$+ P_s(\omega,\cos\theta)(\hat{\underline{\mu}}(\omega)\cdot\underline{e}_\phi)\underline{e}_\phi\} \quad , \tag{9.15}$$

where we have used that $P_\sigma(\omega,\cos\theta)$ is only nonzero in a small frequency band around $\bar{\omega}$.

X. NARROW-BAND EXCITATION

The Fresnel coefficients $R_\sigma(\omega,\cos\theta)$ and $P_\sigma(\omega,\cos\theta)$ depend in general in a complicated way on the frequency ω. For R_σ this is mainly brought about by the variation of the dielectric constant with ω, but P_σ has an additional geometric frequency dependence. The value of P_σ has a sharp peak around $\omega \approx \bar{\omega}$, and the relative frequency width of the response is on the order of $|\rho-1| \sim \gamma$. The absolute frequency width then becomes $|\omega-\bar{\omega}| \sim \gamma\bar{\omega}$, and even for very small interaction parameters γ this can still be very large due to the multiplication by $\bar{\omega}$. We now assume that the dipole radiation is nearly monochromatic, compared to the frequency width $\gamma\bar{\omega}$ of the PC. With ω_o as the central frequency of the dipole field and with $\Delta\omega$ as its typical width, we impose the restriction $\Delta\omega \ll \gamma\bar{\omega}$ on the exciting field. However, this does not imply that ω_o has to be in close resonance with $\bar{\omega}$, but only that the spectral

width of the dipole radiation is small compared to $\gamma\bar{\omega}$. For an atomic transition between (nearly) degenerate states which are separated by ω_o, this condition is very easily met.

Under the above condition we can replace $R_\sigma(\omega,\cos\theta)$ and $P_\sigma(\omega,\cos\theta)$ by their values at $\omega = \omega_o$. Furthermore, the factor ω^2 in the integrands can be taken outside the integrals as ω_o^2. The only remaining frequency dependence of the integrands enters as $\hat{\underset{\sim}{\mu}}(\omega)$ or its mirror image, and the ω-integrals can be performed easily. We find for the three fields

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = \frac{\omega_o^2}{4\pi\epsilon_o rc^2}\{(\underset{\sim}{\mu}(t-r/c+\tau)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta + (\underset{\sim}{\mu}(t-r/c+\tau)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{10.1}$$

$$\underset{\sim}{E}_r(\underset{\sim}{r},t)^{(+)} = \frac{\omega_o^2}{4\pi\epsilon_o rc^2}\{R_p(\omega_o,\cos\theta)(\underset{\sim}{\mu}'(t-r/c-\tau)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta - R_s(\omega_o,\cos\theta)(\underset{\sim}{\mu}'(t-r/c-\tau)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{10.2}$$

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = -\frac{\omega_o^2}{4\pi\epsilon_o rc^2}\, e^{2i\bar{\omega}(t-r/c+\tau)} \times \{P_p(\omega_o,\cos\theta)(\underset{\sim}{\mu}(t-r/c+\tau)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta + P_s(\omega_o,\cos\theta)(\underset{\sim}{\mu}(t-r/c+\tau)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{10.3}$$

in terms of the positive-frequency part $\underset{\sim}{\mu}(t)^{(+)}$ of the dipole moment operator.

XI. RETARDATION

The three fields, evaluated at the position $\underline{r}$ of the detector and at time t, are determined by the instantaneous value of the dipole moment at an earlier time $t - r/c \pm \tau$. Since photons travel with the speed of light c, the delay time r/c equals the propagation time of a photon when it would travel from the origin of the coordinate system to the detector. For both the p-wave and the pc-wave, this retardation time is reduced by an amount $\tau = (h/c)\cos\theta$ for a given observation angle θ, and Fig. 2 illustrates that this implies that the photons are emitted from the site of the dipole, and directly into the direction of the detector. The p-wave corresponds to emitted radiation by the dipole into the region $z > h$ without interference from the medium (Sec. IV), which makes this picture consistent. The phase-conjugated wave, however, is a reflected field by the surface, but nevertheless the photons seem to emanate from the site of the dipole and travel directly into the direction of the detector, in view of their retardation time. The specular r-wave has a retardation of 2τ, as compared to the p-wave and the pc-wave, and it is shown in Fig. 3 that this time delay accounts for the difference in travel time between a directly-emitted photon and a photon which is first reflected by the surface $z = 0$. The figure also illustrates that upon reflection at point A the path of a photon obeys Snell's law, and that a photon appears to be emitted by a mirror dipole $\underline{\mu}'$ which is a distance h below the surface. We conclude that the emission of both p- and r-photons is in strong analogy with classical ray optics, and that the quantum nature of the radiation affects neither the geometry nor the interpretation of the light emission. This is in contrast to the emission of the pc-wave. If this wave would be a result of the emission of a photon by the dipole into the direction of the surface, and a subsequent reflection as a phase-conjugated image, then the delay time would

be $2h/\cos\theta$, as compared to direct emission. Since there is no such retardation, we conclude that the emission of phase-conjugated fluorescence is more complicated. We shall show in Sec. XVII how the mechanism of pc-wave emission can be understood.

XII. TOTAL SOURCE FIELD

In this section we construct the total source field $\underset{\sim}{E}_s$, as it is measured by the detector. We shall suppress the overall time delay with r/c. Then, for an atom which is not more than a few wavelengths away from the surface $z = 0$, the time delay τ is only a few optical cycles, and we can safely replace the time evolution of $\underset{\sim}{\mu}(t)$ by its free evolution over this small time interval. This amounts to the approximation

$$\underset{\sim}{\mu}(t+\tau)^{(+)} \simeq e^{-i\omega_o\tau}\underset{\sim}{\mu}(t)^{(+)} \quad , \tag{12.1}$$

which is perfectly justified at optical frequencies. We then obtain for the fields

$$\underset{\sim}{E}_p(\underset{\sim}{r},t)^{(+)} = \frac{\omega_o^2 e^{-i\omega_o\tau}}{4\pi\epsilon_o rc^2}\{(\underset{\sim}{\mu}(t)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta + (\underset{\sim}{\mu}(t)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{12.2}$$

$$\underset{\sim}{E}_r(\underset{\sim}{r},t)^{(+)} = \frac{\omega_o^2 e^{i\omega_o\tau}}{4\pi\epsilon_o rc^2}\{R_p(\omega_o,\cos\theta)(\underset{\sim}{\mu}'(t)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta$$

$$- R_s(\omega_o,\cos\theta)(\underset{\sim}{\mu}'(t)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{12.3}$$

$$\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(-)} = -\frac{\omega_o^2 e^{-i\omega_o\tau}}{4\pi\epsilon_o rc^2} e^{2i\bar{\omega}(t+\tau)}$$

$$\times \{P_p(\omega_o,\cos\theta)(\underset{\sim}{\mu}(t)^{(+)}\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta$$

$$+ P_s(\omega_o,\cos\theta)(\underset{\sim}{\mu}(t)^{(+)}\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{12.4}$$

in which all dipole moments appear with the same time argument t.

Subsequently, we take the Hermitian conjugate of Eq. (12.4) which gives us $\underset{\sim}{E}_{pc}(\underset{\sim}{r},t)^{(+)}$ according to Eq. (2.7), and we use $(\underset{\sim}{\mu}(t)^{(+)})^\dagger = \underset{\sim}{\mu}(t)^{(-)}$. Then we add the three fields, which gives us for the positive-frequency part of the source field

$$\underset{\sim}{E}_s(\underset{\sim}{r},t)^{(+)} = \frac{\omega_o^2 e^{-i\omega_o\tau}}{4\pi\epsilon_o rc^2}\{(\underset{\sim}{M}_\theta(t,\cos\theta)\cdot\underset{\sim}{e}_\theta)\underset{\sim}{e}_\theta + (\underset{\sim}{M}_\phi(t,\cos\theta)\cdot\underset{\sim}{e}_\phi)\underset{\sim}{e}_\phi\} \quad , \tag{12.5}$$

where we introduced the two operators

$$\underset{\sim}{M}_\theta(t,\cos\theta) = \underset{\sim}{\mu}(t)^{(+)} + e^{2i\omega_o\tau}\{R_p(\omega_o,\cos\theta)\underset{\sim}{\mu}'(t)^{(+)}$$

$$- e^{-2i\bar{\omega}(t+\tau)}P_p(\omega_o,\cos\theta)^*\underset{\sim}{\mu}(t)^{(-)}\} \quad , \tag{12.6}$$

$$\underset{\sim}{M}_\phi(t,\cos\theta) = \underset{\sim}{\mu}(t)^{(+)} - e^{2i\omega_o\tau}\{R_s(\omega_o,\cos\theta)\underset{\sim}{\mu}'(t)^{(+)}$$

$$+ e^{-2i\bar{\omega}(t+\tau)}P_s(\omega_o,\cos\theta)^*\underset{\sim}{\mu}(t)^{(-)}\} \quad . \tag{12.7}$$

Without the presence of the medium both operators reduce to $\underset{\sim}{M}_\theta = \underset{\sim}{M}_\phi = \underset{\sim}{\mu}(t)^{(+)}$, and conversely we can interpret $\underset{\sim}{M}_\vartheta$ and $\underset{\sim}{M}_\phi$ as two components of the positive-frequency part of an effective dipole operator which takes into account the interaction with the medium. Notice that both operators contain only positive frequencies, due to the appearance of the factors $\exp(-2i\bar{\omega}t)$.

XIII. INTENSITY

When a detector at a distance r and under solid angle $\Omega = (\theta,\phi)$ measures an electric field $\underset{\sim}{E}_s$, then the detected energy at time t per unit of time and per unit solid angle is in general given by[39]

$$\frac{\partial^2 W}{\partial t \partial \Omega} = 2\epsilon_o c r^2 \langle \underset{\sim}{E}_s(\underset{\sim}{r},t)^{(-)} \cdot \underset{\sim}{E}_s(\underset{\sim}{r},t)^{(+)} \rangle \quad , \tag{13.1}$$

provided that the efficiency is 100%. Here the angle brackets indicate a quantum expectation value. The emitted power into the half-space $z > 0$ is defined by

$$\frac{dW}{dt} = \int_{\substack{\text{half sphere} \\ \text{in } z > 0}} d\Omega \, \frac{\partial^2 W}{\partial t \partial \Omega} \quad . \tag{13.2}$$

With expression (12.5) for the source field, we obtain for the intensity distribution

$$\frac{\partial^2 W}{\partial t \partial \Omega} = \frac{\omega_o^4}{8\pi^2 \epsilon_o c^3} \langle (\underset{\sim}{M}_\theta(t,\cos\theta)^\dagger \cdot \underset{\sim}{e}_\theta)(\underset{\sim}{M}_\theta(t,\cos\theta) \cdot \underset{\sim}{e}_\theta)
$$

$$+ (\underset{\sim}{M}_{\phi}(t,\cos\theta)^{\dagger}\cdot\underset{\sim}{e}_{\phi})(\underset{\sim}{M}_{\phi}(t,\cos\theta)\cdot\underset{\sim}{e}_{\phi})> \quad , \tag{13.3}$$

and substitution of the explicit forms of $\underset{\sim}{M}_{\theta}$ and $\underset{\sim}{M}_{\phi}$ then gives the angular radiation pattern.

Both operators $\underset{\sim}{M}_{\theta}$ and $\underset{\sim}{M}_{\phi}$ depend only on the angle θ, and therefore the ϕ-dependence in $\partial^2 W/\partial t\partial\Omega$ is purely geometrical, which reflects the invariance of the system for rotations about the z-axis. Integration over ϕ and θ then yields for the emitted power

$$\frac{dW}{dt} = a(b_{\parallel}<\underset{\sim}{\mu}(t)_{\parallel}^{(-)}\cdot\underset{\sim}{\mu}(t)_{\parallel}^{(+)}> + b_{\perp}<\underset{\sim}{\mu}(t)_{\perp}^{(-)}\cdot\underset{\sim}{\mu}(t)_{\perp}^{(+)}>$$

$$+ c_{\parallel}<\underset{\sim}{\mu}(t)_{\parallel}^{(+)}\cdot\underset{\sim}{\mu}(t)_{\parallel}^{(-)}> + c_{\perp}<\underset{\sim}{\mu}(t)_{\perp}^{(+)}\cdot\underset{\sim}{\mu}(t)_{\perp}^{(-)}>) \quad , \tag{13.4}$$

where the overall parameter is given by

$$a = \frac{\omega_o^4}{6\pi\epsilon_o c^3} \quad , \tag{13.5}$$

and the parameter functions are

$$b_{\parallel} = \frac{3}{4}\int_0^1 du(|1 + e^{i\beta u}R_s(\omega_o,u)|^2 + u^2|1 - e^{i\beta u}R_p(\omega_o,u)|^2) \quad , \tag{13.6}$$

$$b_{\perp} = \frac{3}{2}\int_0^1 du(1-u^2)|1 + e^{i\beta u}R_p(\omega_o,u)|^2 \quad , \tag{13.7}$$

$$c_{\parallel} = \frac{3}{4}\int_0^1 du(|P_s(\omega_o,u)|^2 + u^2|P_p(\omega_o,u)|^2) \quad , \tag{13.8}$$

$$c_{\perp} = \frac{3}{2}\int_0^1 du(1-u^2)\left|P_p(\omega_o,u)\right|^2 \quad . \tag{13.9}$$

Here, u signifies the cosine of the angle of incidence of a plane wave, and the integrals represent the superposition of plane waves. The parameter functions $b_{\parallel}$, $b_{\perp}$, $c_{\parallel}$ and $c_{\perp}$ are real and positive, and they are determined by the Fresnel reflection coefficients. These four parameters incorporate entirely all the properties of the medium, like its dielectric constant, nonlinear interaction parameter and geometry (for instance, the layer thickness), and they are independent of the dipole moment $\underset{\sim}{\mu}$. Therefore, the representation (13.4) for the emission rate makes a clear separation between material properties and the dynamic evolution of the atom, which is incorporated in the time dependence of the dipole moment $\underset{\sim}{\mu}(t)$. This result is reminiscent of the general form of the spontaneous-decay operator for an atom near a linear medium, which can also be expressed in terms of the parameter functions $b_{\parallel}$ and $b_{\perp}$ and dipole-moment expectation values.[40] The connection is, of course, that spontaneous emission and spontaneous decay are intimately related.

The parameter β in Eqs. (13.6) and (13.7) is defined as

$$\beta = 2\omega_o h/c \quad , \tag{13.10}$$

which equals 4π times the distance h, measured in optical wavelengths. This parameter enters through the $\cos\theta$-dependence of the delay time τ, and the factor $\exp(i\beta u)$ accounts for the proper phase relation between the interfering p-waves and r-waves. There is no such factor in the expressions for $c_{\parallel}$ and $c_{\perp}$ (which describe the pc-wave emission), and this indicates again that the

phase-conjugated fluorescence is of a different nature than ordinary fluorescence (p-waves and r-waves). Especially, the pc-waves do not interfere with the other components of the emitted radiation. Also notice that the parameters $b_{\parallel}$ and $b_{\perp}$ depend on the normal distance between the atom and the surface (through β), but that $c_{\parallel}$ and $c_{\perp}$ are independent of h. It can be shown that this is a consequence of approximation (12.1), which limits the range of h to a few optical wavelengths. In physical terms, the atom does not decay on a time scale τ.

XIV. PARAMETER FUNCTIONS

In order to shed some light on the significance of the parameter functions, we consider some examples.

A. Transparent medium

When the dielectric constant ϵ of the medium equals unity and the nonlinear interaction parameter γ equals zero, then there is effectively no medium at all, which gives

$$R_s = R_p = P_s = P_p = 0 \tag{14.1}$$

for the Fresnel coefficients. Then the parameter functions are found to be

$$b_{\parallel} = b_{\perp} = 1 \quad , \quad c_{\parallel} = c_{\perp} = 0 \quad . \tag{14.2}$$

Consequently, a deviation of $b_{\parallel}$ or $b_{\perp}$ from unity reflects the presence of specular waves, and a nonvanishing $c_{\parallel}$ or $c_{\perp}$ indicates a phase-conjugate signal. The emitted power becomes

$$\frac{dW}{dt} = a \langle \underset{\sim}{\mu}(t)^{(-)} \cdot \underset{\sim}{\mu}(t)^{(+)} \rangle \quad . \tag{14.3}$$

B. Perfect conductor

For the case of a perfect-conducting medium (mirror) the parameter functions can readily be evaluated. The Fresnel coefficients are

$$P_s = P_p = 0 \quad , \quad R_s = -1 \quad , \quad R_p = 1 \quad , \tag{14.4}$$

which gives of course

$$c_{\parallel} = c_{\perp} = 0 \quad . \tag{14.5}$$

For the coefficients of linear reflection we find

$$b_{\parallel} = 2 - 3\left(\frac{\sin\beta}{\beta} + \frac{\cos\beta}{\beta^2} - \frac{\sin\beta}{\beta^3}\right) \quad , \tag{14.6}$$

$$b_{\perp} = 2 - 6\left(\frac{\cos\beta}{\beta^2} - \frac{\sin\beta}{\beta^3}\right) \quad , \tag{14.7}$$

which is a well-known result.[41,42]

C. Ideal PC

For a plane wave at normal incidence, the distinction between s-waves and p-waves disappears, and we have $P_s = P_p$. When most of the incident radiation is normal to the surface, we can make the approximation $P_s \approx P_p$ and take the value $u = 1$. For an ideal PC the reflection coefficient is

independent of the polarization and the angle of incidence, and there is no specular reflection. Then the Fresnel coefficients are

$$R_s = R_p = 0 \quad , \quad P_s = P_p = P \quad , \tag{14.8}$$

and in analogy with the values for a perfect conductor, Eq. (14.4), such a device is sometimes called a phase-conjugating mirror. We find

$$b_{\parallel} = b_{\perp} = 1 \quad , \quad c_{\parallel} = c_{\perp} = |P|^2 \quad , \tag{14.9}$$

and the emission rate becomes

$$\frac{dW}{dt} = a(<\underset{\sim}{\mu}(t)^{(-)} \cdot \underset{\sim}{\mu}(t)^{(+)}> + |P|^2 <\underset{\sim}{\mu}(t)^{(+)} \cdot \underset{\sim}{\mu}(t)^{(-)}>) \quad . \tag{14.10}$$

D. Transparent PC on resonance

A transparent medium is defined as having a unit dielectric constant, which gives for the parameters of the directly-emitted and specular waves

$$R_s = R_p = 0 \quad , \quad b_{\parallel} = b_{\perp} = 1 \quad . \tag{14.11}$$

If we assume that the atomic transition frequency ω_o is in close resonance with the pump fields, e.g.,

$$|\omega_o - \bar{\omega}| \ll \gamma\bar{\omega} \quad , \tag{14.12}$$

then the Fresnel reflection coefficients take the form[43]

$$|P_s(\omega_o,u)|^2 = \tan^2(\frac{\eta}{u}) \quad , \tag{14.13}$$

$$|P_p(\omega_o,u)|^2 = \tan^2(\frac{\eta}{u}(3-2u^2)) \quad , \tag{14.14}$$

in terms of the dimensionless parameter

$$\eta = \gamma \frac{\Delta\omega_o}{2c} \quad , \tag{14.15}$$

where Δ is the layer thickness of the medium. For almost perpendicular incidence these expressions reduce to $|P_s|^2 = |P_p|^2 = \tan^2\eta$, and this behavior has been confirmed qualitatively by experiment.[44,45] It is also well known that for certain values of η (or Δ and ω_o) the magnitude of $|P_s|^2$ or $|P_p|^2$ can become infinite, as follows from Eqs. (14.13) and (14.14). This phenomenon is termed self-oscillation and has been observed in experiment also. For the present problem, however, the cosine of the angle of incidence is a variable, rather than the parameter η, and it is seen from Eq. (14.13) that for

$$\cos\vartheta = \frac{\eta}{(n+\frac{1}{2})\pi} \quad , \tag{14.16}$$

with n integer, the value of $|P_s|^2$ is also infinite. In fact, condition (14.16) predicts an infinite series of values of ϑ in the range (0°,90°) for which $|P_s|^2$ diverges. In a similar way, the reflectivity $|P_p|^2$ has a series of resonances for certain angles of incidence.

When we substitute the expressions (14.13) and (14.14) into Eqs. (13.8) and (13.9), then it is easy to verify that the integrals over u for $c_\parallel$ and $c_\perp$ diverge. This yields an infinite emission rate for the phase-conjugated

fluorescence, which is of course unphysical. It can be shown,[41] however, that when we allow for a finite but very small detuning $\omega_o - \bar{\omega} \neq 0$, the Fresnel coefficients remain finite, with a magnitude on the order of unity. This renders finite values for the parameters $c_\parallel$ and $c_\perp$, and thereby for the mission rate. For an extremely close resonance between ω_o and $\bar{\omega}$ we can no longer replace $P_\sigma(\omega,\cos\theta)$ in Eq. (9.15) by $P_\sigma(\omega_o,\cos\theta)$, and we have to take into account the frequency dependence of $P_\sigma(\omega,\cos\theta)$ over the width of the atomic emission line.

This situation is analogous to the problem of resonance fluorescence by a two-level atom in a monochromatic laser field. There, the spectral distribution of the fluorescence consists of a δ-function at the laser frequency, superposed on a smooth background, and in the limit where ω_o equals the laser frequency the emission rate is infinite. In any case, the example of this section shows that the situation of perfect resonance has to be considered with caution.

XV. SCHRÖDINGER PICTURE

The time dependence of the emission rate dW/dt in Eq. (13.4) is governed by the time evolution of the Heisenberg operator $\underline{\mu}(t)$. A more transparent representation of dW/dt can be obtained by a transformation to the Schrödinger picture. When we take the Schrödinger and Heisenberg picture to coincide at $t = 0$, then we have

$$\underline{\mu}(0) = \underline{\mu} = \underline{\mu}^{(+)} + \underline{\mu}^{(-)} \quad , \tag{15.1}$$

with

$$\underline{\mu}^{(+)} = P_g \underline{\mu} P_e \quad , \tag{15.2}$$

$$\underline{\mu}^{(-)} = P_e \underline{\mu} P_g \quad , \tag{15.3}$$

in terms of the projectors P_e and P_g onto the (possibly degenerate) excited and ground levels, respectively. In free evolution (no coupling to the radiation field), the Heisenberg representation of the lowering operator $\underline{\mu}^{(+)}$ is $\underline{\mu}^{(+)}(t) = \exp(-i\omega_o t)\underline{\mu}^{(+)}$, which equals the positive-frequency part of $\underline{\mu}(t)$, e.g., $\underline{\mu}(t)^{(+)} = \underline{\mu}^{(+)}(t)$. Due to the interaction with the electromagnetic field, the spectral distribution of $\underline{\mu}^{(+)}(t)$ will acquire a finite width around the central frequency ω_o, but to an excellent approximation this operator will still contain positive frequencies only. Hence we can set

$$\underline{\mu}(t)^{(\pm)} = \underline{\mu}^{(\pm)}(t) \quad . \tag{15.4}$$

For any two operators A(t) and B(t) we have the identity

$$\langle A(t)B(t)\rangle = \mathrm{Tr}\rho(t)A(0)B(0) \quad , \tag{15.5}$$

in terms of the density operator $\rho(t)$ of the entire atom plus field system. In Eq. (13.4), A(t) and B(t) are Cartesian components of the dipole operator, and in the Schrödinger picture they act on wave functions in the atomic Hilbert space only. Then we can take the trace over the field states in Eq. (15.5) according to

$$\rho_a(t) = \mathrm{Tr}_{\mathrm{field}}\rho(t) \quad , \tag{15.6}$$

where $\rho_a(t)$ is the reduced density operator for the atom, irrespective of the state of the field. In this way, we find for the emission rate

$$\frac{dW}{dt} = a\ \mathrm{Tr}_a \rho_a(t)\{b_{\|}\underline{\mu}_{\|}^{(-)}\cdot\underline{\mu}_{\|}^{(+)} + b_{\perp}\underline{\mu}_{\perp}^{(-)}\cdot\underline{\mu}_{\perp}^{(+)}$$

$$+ c_{\|}\underline{\mu}_{\|}^{(+)}\cdot\underline{\mu}_{\|}^{(-)} + c_{\perp}\underline{\mu}_{\perp}^{(+)}\cdot\underline{\mu}_{\perp}^{(-)}\} \quad , \tag{15.7}$$

where the trace runs over the atomic states only. Equation (15.7) expresses that we can find the emission rate at time t, once the atomic density operator $\rho_a(t)$ is known, since the operator in braces is completely determined by Eqs. (15.2) and (15.3).

XVI. TWO-STATE ATOM AND IDEAL PC

In order to exhibit the principle features of the emission of phase-conjugated fluorescence, we work out the example of a two-state atom, in combination with the model (ideal) PC from Sec. XIV.C. A model two-state atom has an excited state $|e\rangle$ and a ground state $|g\rangle$, and the projectors on these states are $P_e = |e\rangle\langle e|$ and $P_g = |g\rangle\langle g|$, respectively. Then it is easy to work out the emission rate dW/dt from Eq. (15.7), and we obtain

$$\frac{dW}{dt} = \frac{1}{2} A\hbar\omega_o\{n_e(t) + |P|^2 n_g(t)\} \quad , \tag{16.1}$$

where

$$A = \frac{\omega_o^3}{3\pi\epsilon_o\hbar c^3}\,|\langle e|\underline{\mu}|g\rangle|^2 \tag{16.2}$$

equals the Einstein coefficient for spontaneous emission by an atom in empty space. The time dependence of dW/dt enters through the populations of the atomic levels, defined by

$$n_i(t) = \mathrm{Tr}_a \rho_a(t) P_i \quad , \quad i = e,g \quad . \tag{16.3}$$

These quantities cannot be determined with the present theory, since the time evolution of $\rho_a(t)$ is governed by the coupling of the atomic dipole to the vacuum field (and possible external fields). Also, $\rho_a(t)$ depends on the preparation of the system at $t = 0$. Nevertheless, when for a given time t the populations are $n_e(t)$ and $n_g(t)$, then the emission rate follows from Eq. (16.1). This situation is analogous to the fact that for fluorescent emission by a dipole in empty space, the emission rate always equals $dW/dt = A\hbar\omega_o n_e(t)$, no matter how the excited state became populated.

The emission rate has two distinct contributions. The first term is $\frac{1}{2}A\hbar\omega_o n_e(t)$, which equals half the emission rate in empty space. These are the photons that are emitted directly by the atom towards the detector (p-wave), and the factor $\frac{1}{2}$ accounts for the fact that we only measure the emission into the half-space $z > 0$. This contribution is always present for any medium in $z < 0$, simply because this part of the field does not interact with the medium. During the emission the atom decays from its excited state to the ground state as illustrated in Fig. 4.a. The other half of the number of photons which are emitted in the $|e\rangle \rightarrow |g\rangle$ transition travel in the negative z-direction, and they serve as the incident field on the medium. For linear reflection this would give rise to a specular wave, with intensity proportional to $n_e(t)$. Interference between the p-wave and the r-wave is incorporated in the values of the parameter functions $b_\parallel$ and $b_\perp$.

More interesting is the second contribution $\frac{1}{2}A\hbar\omega_o|P|^2 n_g(t)$ to the emission rate. This term is proportional to $|P|^2$ and represents therefore the phase-conjugated fluorescence, as can be measured by a detector. The question to ask is what physical mechanism is responsible for the emission of this radiation. As pointed out in Sec. XI, an interpretation in terms of an incident wave which is reflected as a pc-wave cannot be correct, since this is in conflict with the different retardation times of the various photons. First, we notice that this term corresponds to the actual observation of a photon, and as we found from the retardation time, this photon is emitted from the site of the atom. This emission can only take place if accompanied by an $|e\rangle \rightarrow |g\rangle$ atomic transition. On the other hand, the emission rate is proportional to $n_g(t)$ so that the atom must be originally in its ground state. Consequently, before the emission/decay process the atom must be excited, and because of energy conservation this can only happen in combination with photon absorption. The only photon source in the problem, assuming no external fields, is the PC. Therefore, we must conclude that this photon is spontaneously emitted by the medium, due to the presence of the dipole. Such a process would give rise to the emission of a photon with frequency $\bar{\omega}$, but we recall that this phase-conjugated wave must have frequency $2\bar{\omega}-\omega_o$, since the incident field has frequency ω_o. An energy-conserving process then requires that a second photon with frequency $\bar{\omega}$ is absorbed by the atom, which gives rise to an excitation from $|g\rangle$ to $|e\rangle$. Figure 4.b illustrates this three-photon process.

XVII. DEGENERATE TWO-LEVEL ATOM

A two-state atom description is an idealization which is commonly employed to study the principles of matter-radiation interactions. In this

fashion we can discover the fundamental mechanisms of a certain process, such as illustrated in Fig. 4 for the present problem, although the model is not necessarily very realistic. In this section we consider a degenerate two-level atom, where the excited and ground levels have angular momenta j_e and j_g, respectively. These levels are $(2j_e+1)$- and $(2j_g+1)$-fold degenerate, and the magnetic states are indicated by $|j_e m_e\rangle$ and $|j_g m_g\rangle$ in obvious notation. Then it is immediately clear that a two-state description cannot account for the behavior of the atom, even if one would select a certain transition between two states by an external field. For instance, in a $j_g = 0, j_e = 1$ system we can select the $m_e = m_g = 0$ states by driving the atom with a linearly-polarized laser in the z-direction. Stimulated transitions occur only between the $|0\ 0\rangle$ and $|1\ 0\rangle$ states, and in addition we have the two spontaneous processes from Fig. 4 between these states. But then also the $|1\ 1\rangle$ and $|1\ -1\rangle$ excited states have a nonzero dipole matrix element with the ground state $|0\ 0\rangle$, and the three-photon loops from Fig. 4.b will generate fluorescence whenever the atom is in its ground state.

In this section we evaluate the general expression for the emission rate for a degenerate two-level atom. In terms of the dipole-selective raising operator

$$d_\tau = \sum_{m_e m_g} (j_g m_g 1\tau | j_e m_e) |j_e m_e\rangle\langle j_g m_g| \quad , \quad \tau = -1,\ 0,\ 1 \quad , \tag{17.1}$$

the lowering part of the dipole operator attains the form

$$\underset{\sim}{\mu}^{(+)} = \frac{\langle j_e || \mu || j_g \rangle^*}{\sqrt{2j_e + 1}} \sum_\tau d_\tau^\dagger \underset{\sim}{e}_\tau \quad . \tag{17.2}$$

in terms of the spherical unit vectors $\underset{\sim}{e}_\tau$ with respect to the z-axis. The summation over $\tau = \pm 1$ then gives $\underset{\sim}{\mu}_{\parallel}^{(+)}$, whereas the $\tau = 0$ term equals the perpendicular part of $\underset{\sim}{\mu}^{(+)}$. With the Einstein coefficient for spontaneous emission in free space and for a degenerate system,

$$A = \frac{\omega_o^3}{3\pi\epsilon_o \hbar c^3} \frac{|\langle j_e||\mu||j_g\rangle|^2}{2j_e + 1} \quad , \tag{17.3}$$

and with the change in notation

$$b_\tau = \begin{cases} b_{\parallel} \quad , & \text{for } \tau = \pm 1 \\ \\ b_{\perp} \quad , & \text{for } \tau = 0 \end{cases} \quad , \tag{17.4}$$

$$c_\tau = \begin{cases} c_{\parallel} \quad , & \text{for } \tau = \pm 1 \\ \\ c_{\perp} \quad , & \text{for } \tau = 0 \end{cases} \quad , \tag{17.5}$$

the emission rate becomes

$$\frac{dW}{dt} = \frac{1}{2} \hbar\omega_o A \mathrm{Tr}_a \rho_a(t) \sum_\tau (b_\tau d_\tau d_\tau^\dagger + c_\tau d_\tau^\dagger d_\tau) \quad . \tag{17.6}$$

In order to demonstrate the similarities and differences with a two-state atom, we consider again the case of an ideal PC, for which $b_\tau = 1$ and $c_\tau = |P|^2$. With the identities

$$\sum_{\tau} d_{\tau} d_{\tau}^{\dagger} = P_e \quad , \tag{17.7}$$

$$\sum_{\tau} d_{\tau}^{\dagger} d_{\tau} = \frac{2j_e + 1}{2j_g + 1} P_g \quad , \tag{17.8}$$

in terms of the projectors onto the two levels, we find

$$\frac{dW}{dt} = \frac{1}{2} \hbar\omega_o A \mathrm{Tr}_a \rho_a(t) \left(P_e + |P|^2 \frac{2j_e + 1}{2j_g + 1} P_g \right) \quad . \tag{17.9}$$

With the notation

$$A_g = \frac{1}{2} \frac{2j_e + 1}{2j_g + 1} A \, |P|^2 \quad , \tag{17.10}$$

this can be written as

$$\frac{dW}{dt} = \frac{1}{2} \hbar\omega_o A n_e(t) + \hbar\omega_o A_g n_g(t) \quad , \tag{17.11}$$

where the populations of the levels are again given by Eq. (16.3). Comparison with Eq. (16.1) shows that the only difference is that the ground-level emission term is now multiplied by the geometrical factor $(2j_e+1)/(2j_g+1)$. Notice that only the total populations of the levels determine the fluorescent emission rate; it does not matter how the populations are distributed over the various states. This holds only for an ideal PC, and not in the more general situation, as given by Eq. (17.6).

XVIII. CONCLUSIONS

We have studied the emission of fluorescence radiation by an atom in the vicinity of a phase-conjugating surface. With a decomposition of the particular solution (dipole in empty space) of the Maxwell-Heisenberg equations into an angular spectrum of plane waves, we have been able to construct the reflected field by the medium. Under the assumption that the PC operates via four-wave mixing, the reflected field could be expressed in terms of the classical Fresnel reflection coefficients R_σ and P_σ. These parameters incorporate the material properties of the medium and the details of the geometry for the four-wave mixing process (like the polarization of the pump beams, and the layer thickness). This procedure yields the exact solution for the electromagnetic field in the entire region $z > 0$ in terms of angular integrals, and with an asymptotic expansion we have derived the value of the field in the far zone, in terms of the observation angle (θ,ϕ). After integration over a 2π solid angle we have obtained the total fluorescent power in the half-space $z > 0$, where it turns out that this power can be expressed in terms of four geometrical parameter functions $b_\parallel$, $b_\perp$, $c_\parallel$ and $c_\perp$, and quantum expectation values of the equal-time autorcorrelation functions of the Cartesian components of the lowering part of the dipole operator. By considering various limiting cases in Sec. XIV, we have shown that the terms proportional to $b_\parallel$ and $b_\perp$ account for the fluorescent emission directly from the atom towards the detector. Also included in these terms is the specular reflection of photons, and it appeared that both waves interfere. This interference is again a pure geometrical and classical phenomenon, and is accounted for by the functions $b_\parallel$ and $b_\perp$. The presence of the phase-conjugated wave appears as a separate term in the expression for the emitted

power, which indicates that this wave does not interfere with the p-waves and the r-waves.

In working out the model case of a two-state atom, we have been able to show that the directly-emitted photons and the specular photons are both produced in a spontaneous decay of the atom. Since both emissions are a result of the same decay process $|e\rangle \rightarrow |g\rangle$, these photons must necessarily interfere, as is expressed by their joint appearance in the single parameter functions $b_{\parallel}$ and $b_{\perp}$. On the other hand, the probability for the emission of a phase-conjugated photon is proportional to the population of the ground state, which renders this mechanism independent of the previous one. By considering the retardation times of the various photons, we can track down the underlying physical mechanisms, and the two responsible processes are illustrated by the diagrams of Fig. 4. The most remarkable result is that an atom in its ground state, and close to a PC, can effectively emit a fluorescent photon, in addition to which the atom excites spontaneously. We have interpreted this phenomenon as a result of a three-photon process, as depited in Fig. 4.b.

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FIGURE CAPTIONS

Fig. 1. The $z = 0$ plane separates the vacuum $z > 0$ from the nonlinear medium in $z < 0$, and the radiating atom is located in $x = y = 0$ and $z = h$. In an angular-spectrum representation, the dipole radiation equals an integral over the parallel components $\underset{\sim}{K}_{\parallel}$ of wave vectors. For $z > h$ and $0 < z < h$ these wave vectors are $\underset{\sim}{K}$ and $\underset{\sim}{K}'$ respectively, and their directions are indicated on the left-hand side of the figure for the case of travelling waves. The $\underset{\sim}{K}$-waves, which are referred to as p-waves in the text, travel directly from the site of the atom in $z = h$ towards the detector in $z >> h$, whereas the waves with wave vector $\underset{\sim}{K}'$ serve as the incident field on the medium. In the case of evanescent waves, the field amplitudes decay in the z-direction as shown pictorially on the right-hand side, and these waves travel in the $\underset{\sim}{K}_{\parallel}$ direction. Also shown is the phase convention for the unit polarization vectors, both for $\underset{\sim}{K}$ and $\underset{\sim}{K}'$ waves.

Fig. 2. The atomic dipole is located at $\underset{\sim}{r} = \underset{\sim}{h}$, and the emitted fluorescence is detected within an angle θ with the z-axis. Both the directly-emitted (p-wave) and phase-conjugated (pc-wave) radiation appears to emanate from the location of the dipole, as follows from the retardation time $\tau = (h/c)\cos\theta$ of the p and pc-waves, and the geometry shown in this figure.

Fig. 3. The retardation time in the observation of a specular wave equals $(2h/c)\cos\theta$, as compared to the observation of a p- or pc-wave. The geometry in this figure shows that this retardation can be interpreted as the delay time for a photon which travels first from the atom to point A, where it is subsequently reflected according to Snell's law in classical optics. Also, the distance $2h\cos\theta$ equals the separation between point B and a mirror dipole $\underset{\sim}{\mu}'$ at a distance h below the surface. For a detector within an angle θ, the

r-photons appear to come from this mirror dipole, as can be seen from the geometry shown in the figure.

Fig. 4. This figure illustrates the two atomic processes which contribute to the observable fluorescence. Diagram (a) represents an atom which decays from its excited state $|e\rangle$ to its ground state $|g\rangle$ under emission of a photon into the direction of the detector. This is the p-wave, and the mechanism is not affected by the presence of the medium. The mechanism for the emission of phase-conjugated fluorescence is shown in diagram (b). The atom is originally in its ground state. Spontaneous emission of a photon (pc-wave) by the PC, and subsequent absorption of this photon by the atom, brings the atom to its excited state. Spontaneous decay is then accompanied by the emission of the observable photon. A second absorption of a photon with frequency $\bar{\omega}$ then completes the process, thereby leaving the atom in its excited state.

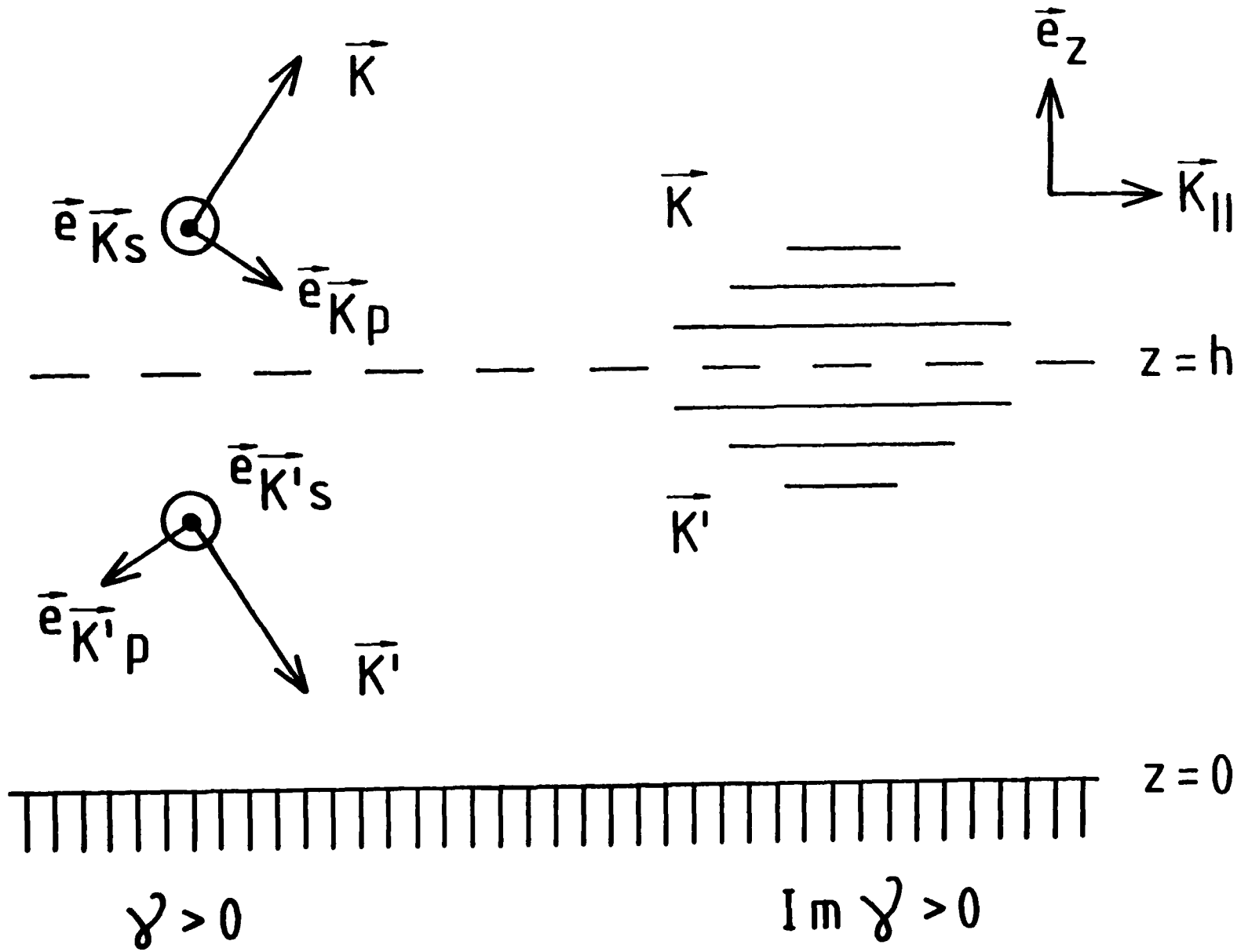

$\vec{K}$
$\vec{e}_{\vec{K}s}$
$\vec{e}_{\vec{K}p}$
$\vec{e}_z$
$\vec{K}_{||}$
$\vec{K}$
z = h
$\vec{e}_{\vec{K}'s}$
$\vec{e}_{\vec{K}'p}$
$\vec{K}'$
$\vec{K}'$
z = 0
$\gamma > 0$
$\mathrm{Im}\,\gamma > 0$

Fig. =

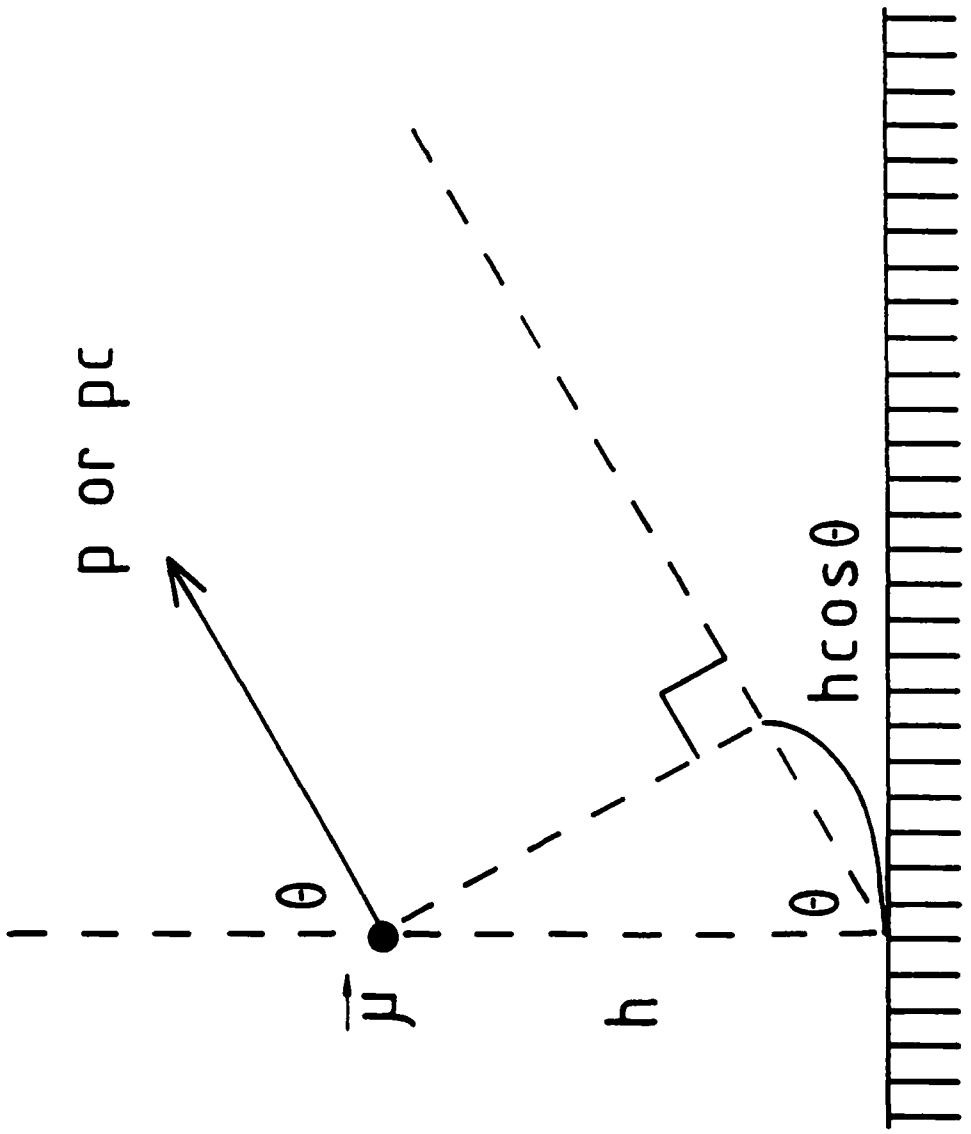
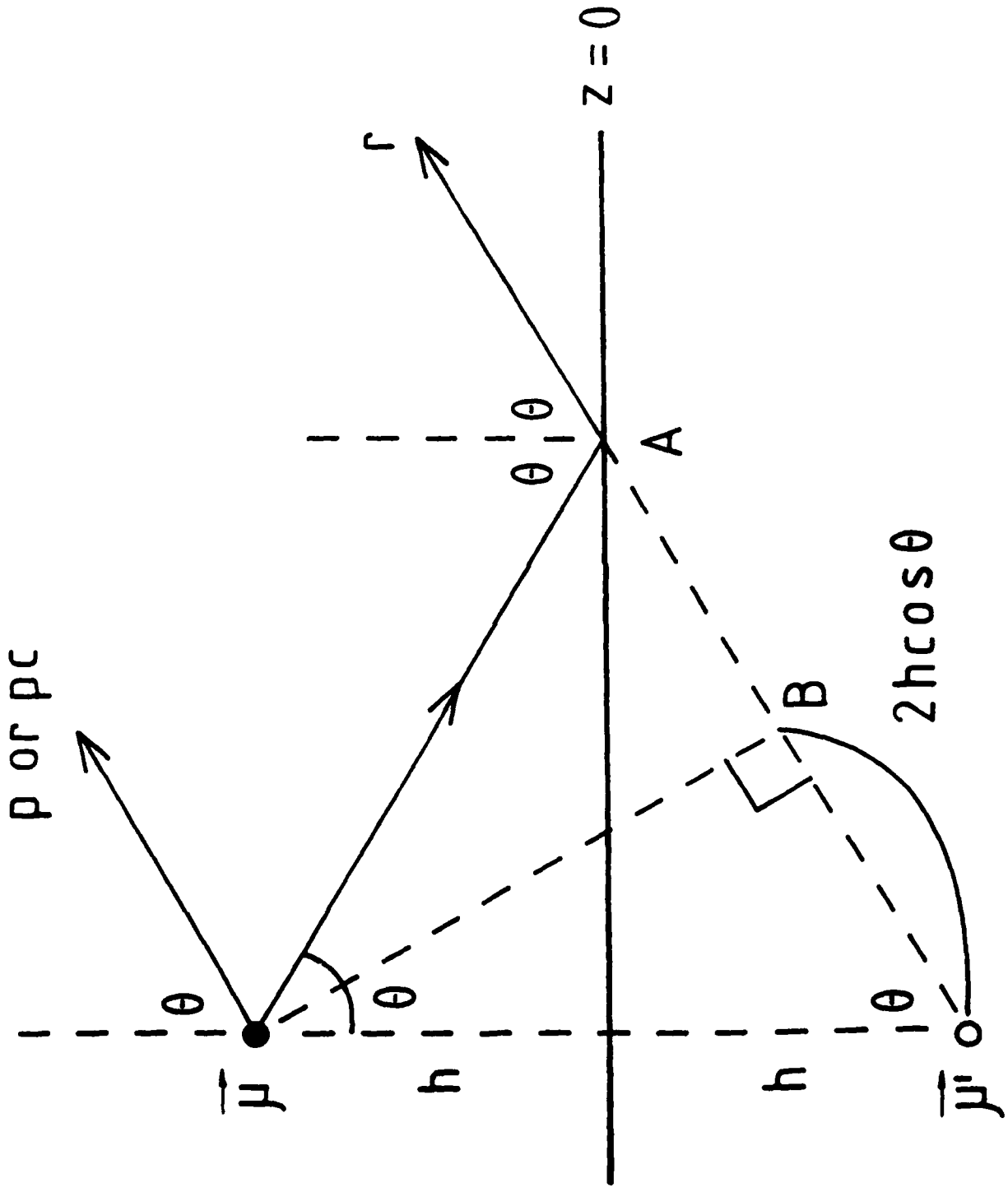
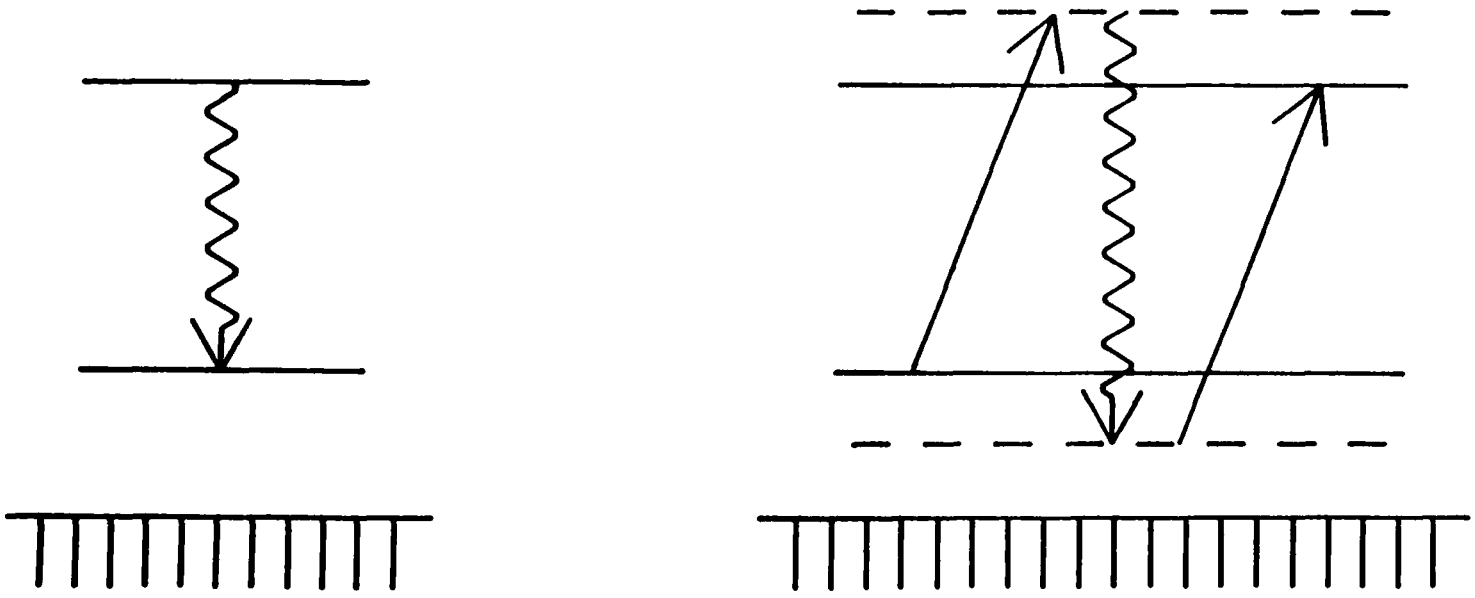


Fig. 3





a

b

Fig. 1

FY90 Abstracts Distribution List for Solid State & Surface Chemistry

Professor John Baldeschwieler
Department of Chemistry
California Inst. of Technology
Pasadena, CA 91125

Professor Paul Barbara
Department of Chemistry
University of Minnesota
Minneapolis, MN 55455-0431

Dr. Duncan Brown
Advanced Technology Materials
520-D Danury Rd.
New Milford, CT 06776

Professor Stanley Bruckenstein
Department of Chemistry
State University of New York
Buffalo, NY 14214

Professor Carolyn Cassady
Department of Chemistry
Miami University
Oxford, OH 45056

Professor R.P.H. Chang
Dept. Matls. Sci. & Engineering
Northwestern University
Evanston, IL 60208

Professor Frank DiSalvo
Department of Chemistry
Cornell University
Ithaca, NY 14853

Dr. James Duncan
Federal Systems Division
Eastman Kodak Company
Rochester, NY 14650-2156

Professor Arthur Ellis
Department of Chemistry
University of Wisconsin
Madison, WI 53706

Professor Mustafa El-Sayed
Department of Chemistry
University of California
Los Angeles, CA 90024

Professor John Eyler
Department of Chemistry
University of Florida
Gainesville, FL 32611

Professor James Garvey
Department of Chemistry
State University of New York
Buffalo, NY 14214

Professor Steven George
Department of Chemistry
Stanford University
Stanford, CA 94305

Professor Tom George
Dept. of Chemistry and Physics
State Universety of New York
Buffalo, NY 14260

Dr. Robert Hamers
IBM T.J. Watson Research Center
P.O. Box 218
Yorktown Heights, N Y 10598

Professor Charles Harris
Department of Chemistry
University of California
Berkeley, CA 94720

Professor John Hemminger
Department of Chemistry
University of California
Irvine, CA 92717

Professor Leonard Interrante
Department of Chemistry
Rensselaer Polytechnic Institute
Troy, NY 12181

Professor Roald Hoffmann
Department of Chemistry
Cornell University
Ithaca, NY 14853

Professor Eugene Irene
Department of Chemistry
University of North Carolina
Chapel Hill, NC 27514

Dr. Sylvia Johnson
SRI International
333 Ravenswood Avenue
Menlo Park, CA 94025

Dr. Zakya Kafafi
Code 6551
Naval Research Laboratory
Washington, DC 20375-5000

Professor Larry Kesmodel
Department of Physics
Indiana University
Bloomington, IN 47403

Professor Max Lagally
Dept. Metal. & Min. Engineeri
University of Wisconsin
Madison, WI 53706

Dr. Stephen Lieberman
Code 522
Naval Ocean Systems Center
San Diego, CA 92152

Professor M.C. Lin
Department of Chemistry
Emory University
Atlanta, GA 30322

Professor Fred McLafferty
Department of Chemistry
Cornell University
Ithaca, NY 14853-1301

Professor Horia Metiu
Department of Chemistry
University of California
Santa Barbara, CA 93106

Professor Larry Miller
Department of Chemistry
University of Minnesota
Minneapolis. MN 55455-0431

Professor George Morrison
Department of Chemistry
Cornell University
Ithaca, NY 14853

rofessor Daniel Neumark
epartment of Chemistry
niversity of California
erkeley, CA 94720

rofessor David Ramaker
epartment of Chemistry
eorge Washington University
ashington, DC 20052

r. Gary Rubloff
BM T.J. Watson Research Center
.O. Box 218
orktown Heights, NY 10598

rofessor Richard Smalley
epartment of Chemistry
ice University
.O. Box 1892
ouston, TX 77251

rofessor Gerald Stringfellow
ept. of Matls. Sci.
& Engineering
niversity of Utah
alt Lake City, UT 84112

rofessor Galen Stucky
epartment of Chemistry
niversity of California
anta Barbara, CA 93106

rofessor H. Tachikawa
epartment of Chemistry
ackson State University
ackson, MI 39217-0510

rofessor William Unertl
ab. for Surface Sci.
& Technology
niversity of Maine
rono, ME 04469

r. Terrell Vanderah
ode 3854
aval Weapons Center
ina Lake, CA 93555

rofessor John Weaver
ept. of Chem. Eng. & Mat. Sci.
niversity of Minnesota
nneapolis, MN 55455

rofessor Brad Weiner
partment of Chemistry
iversity of Puerto Rico
o Piedras, Puerto Rico 00931

Professor Robert Whetten
Department of Chemistry
University of California
Los Angeles, CA 90024

Professor R. Stanley Williams
Department of Chemistry
University of California
Los Angeles, CA 90024

Professor Nicholas Winograd
Department of Chemistry
Pennsylvania State University
University Park, PA 16902

Professor Aaron Wold
Department of Chemistry
Brown University
Providence, RI 02912

Professor Vicki Wysocki
Department of Chemistry
Virginia Commonwealth University
Richmond, VA 23284-2006

Professor John Yates
Department of Chemistry
University of Pittsburg
Pittsburg, PA 15260